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MODEL CALCULATIONS OF THE MIGRATION OF RADIONUCLIDES FROM A REPOSITORY FOR SPENT NUCLEAR FUEL

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Model calculations of the migration of radionuclides from a repository for spent nuclear fuel

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SUMMARY

Release and migration calculations for all important radionuclides which potentially could escape from a repository for spent nuclear fuel have been performed. The calculations have been done for a repository in fissured crystalline rock with models which account for the leaching of the nuclides from the fuel due to dissolution in the moving water and the further transport along the fissures of the rock. Radionuclide sorption retarding the nuclides is accounted for by the concept of matrix diffusion and sorption on the microfissure surfaces. Dispersion and channeling are modeled as Fickian dispersion. Chain decay is accounted for. Calculations have been performed for migration distances of 5, 50 and 100 m. Sorption data and solubility data for oxidizing as well as reducing conditions have been used.

Transport by sorption on colloidal matter in the water is also treated.

In the "central" case the leaching starts after 100.000 years due to the long life time of the canister, but calculations have also been made for a case of some defective canisters where the leaching starts after 100 years. The results are presented as release rates to the biosphere at the various times.

CONTENTS

1. OBJECTIVES

This study is a part of the Swedish KBS-3 project.

The aim of the study is to assess the release to the geosphere and to the biosphere of radioactive material from a repository for spent unreprocessed nuclear fuel.

2. CALCULATION PRINCIPLES

2.1 General description

The release to the biosphere from a repository for unreprocessed nuclear waste has been calculated for four scenarios with different water flow rates and migration lengths.

The release calculations have been performed in two steps.

i The release from the immediate neighbourhood (near field) of a canister, giving a source concentration for the geosphere migration model.
ii The release after passage through a geological medium.

The two transport mechanisms which have been modeled are with the radionuclides in dissolved form or adsorbed on colloidal particles.

Parameter variations that have been performed for some or all of the different scenarios are: different transport mechanisms, time for start of the leach process, reducing- or oxidizing migration conditions and different values of dispersion coefficients.

2.2 Activity release to the geosphere.

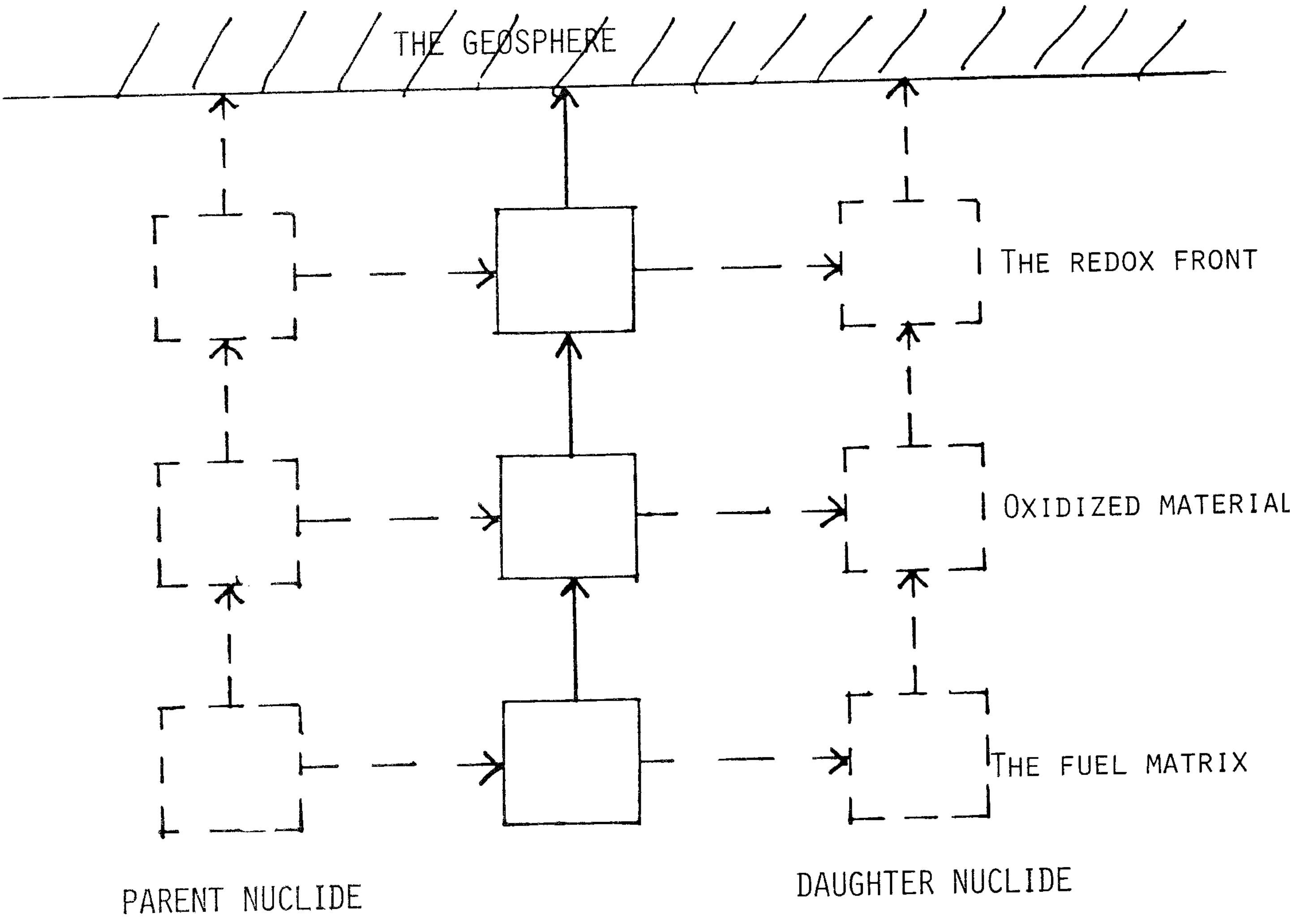
2.2.1 General description

At a certain time the copper-cladding of a canister is assumed to be penetrated as a result of corrosion. Water will then reach the fuel elements and the ionizing radiation will create an oxidizing environment. The oxidizing species (notably hydrogen peroxide and peroxide radicals) will then diffuse out of the canister, through the clay barrier and into the geologic medium together with dissolved fuel constituents in, when so possible, oxidized form.

The rock, at the sites proposed for final disposal of the nuclear waste, contains reducing substances, such as Fe^{2+} which will limit the spreading of the oxidizing zone and lower the oxidation state of some of the migrating species (Neretnieks I,Aslund B, 1983).

As the fuel matrix dissolves its constituents will be released at a rate proportional to the dissolution rate of uranium in oxidized form (congruent dissolution). Eventually the amount of material set free from the matrix may exceed what can be completely dissolved either in oxidized or reduced form.
The interaction between different solid phases affecting the release of a member of a decay chain is schematically depicted in figure 1.

FIGURE 1 INTERACTION BETWEEN DIFFERENT SOLID PHASES IN THE LEACH PROCESS OF A FUEL ELEMENT IN CONTACT WITH WATER.



Further assumptions:

i Homogenous distribution of the fuel constituents except for 10% of the cesium and the iodine which are assumed to be promptly dissolved with a subsequent spread in time of the release caused by a finite rate of replacement of the water in the clay buffer. See further Chapter 5.

ii No coprecipitation.

2.2.2 Release of dissolved material

Release from the fuel matrix

For homogenously distributed nuclides, the release from the fuel matrix can be written as:

$$\frac{M_{i,mat}}{M_{U,tot,mat}} \cdot C_{solub,U,ox} \cdot Q_{eq,ox} \tag{1}$$

As $M_{U,tot,mat} \approx M_{U238,mat}$ and the halflife of U238 in comparison with all the important nuclides, is very long we could approximate $M_{i,Mat}/M_{U,tot,mat}$ by:

$$\frac{M_i^0}{M_{238U}^0} \cdot \text{Decay factor} \tag{2}$$

where the decay factor is the relative change in content of the nuclide i caused by its own decay and by decay of its parent nuclides .

Release from the oxidizing zone

The maximum release from the oxidizing zone of the substance j can be written as:

$$C_{solub,j,ox} \cdot Q_{eq,ox} \quad \text{(grams/year,canister)} \qquad (3)$$

The actual release from the oxidizing zone will be given by (3) provided that the substance exists as a separate solid phase in oxidized form or the rate of release from the fuel matrix plus formation rate of the substance by decay of parent nuclides co-present as separate oxidized phase is larger than the solubility limit given by (3) In that case the release of the isotope i of the substance j will be:

$$\frac{M_{i,ox}}{\sum_j M_{i,ox}} \cdot C_{solub,j,ox} \cdot Q_{eq,ox} \qquad \text{(grams/year,canister)} \qquad (4)$$

Otherwise it will be equal to the release from the fuel matrix plus the formation rate from decay of parent nuclides in the separate solid oxidized phase.

Release from the redox front

The maximum release from the redox front of the substance j can be written as:

$$C_{solub,j,red} \cdot Q_{eq,red} \qquad \text{(grams/year,canister)} \qquad (5)$$

The actual release from the redox front can, in analogy with that from the oxidizing zone, be calculated as the minimum of:

$$\frac{M_{i,red}}{\sum_j M_{i,red}} \cdot C_{solub,j,red} \cdot Q_{eq,red} \qquad \text{(grams/year,canister)} \qquad (6)$$

and the actual release rate from the oxidising zone plus the formation rate from decay of parent nuclides in the separate solid reduced phase.

2.2.3 Release in colloidal form

An estimate of the outflow of radionuclides, adsorbed on colloidal particles, suspended in the water flowing past a canister, has been based on the following premises.

i The adsorption process is assumed to be irreversible.

ii The amount of substance adsorbed is assumed to be given by:

$$K_d \cdot C_{coll} \cdot Q_{eq} \cdot C_i \qquad \text{(grams/year,canister)} \qquad (7)$$

iii The actual release from the near field is given as the maximum of the hypothetical release from the oxidizing zone or from the redox front.

2.3 Activity release to the biosphere

2.3.1 Migration in dissolved form.

The geological medium in which the migration is thought to take place is modelled as cubical blocks of porous rock matrix into which constituents of the surrounding solution diffuses. The dimensions of these blocks are the same as the mean distance between fractures in the rock modelled.

In the calculation of the diffusion into the rock matrix the cubical blocks are approximated with spheres with the same surface to volume ratio as the cubical blocks.

The flowing water is thought to be confined to the interstices between the blocks as the permeability in the rock matrix is very low.

The transport is modelled as one dimensional. The only transverse transport is into the rock matrix.

The transport mechanisms in the fractures will be convective transport and hydrodynamic dispersion modelled as Fickian diffusion with a dispersion coefficient D_L given by a Peclet number Pe:

$$Pe = \frac{U.L}{D_L} \qquad (8)$$

The equation describing the transport of the i-th member in a radionuclide chain will be:

$$\frac{\partial c_f^i}{\partial t} + u_f \frac{\partial c_f^i}{\partial z} - D_L \frac{\partial^2 c_f^i}{\partial z^2} = \alpha N^i - \lambda^i c_f^i + \lambda^{i-1} c_f^{i-1} \qquad (9)$$

$$K^i \frac{\partial c_p^i}{\partial t} = D_p \varepsilon_p \frac{\partial^2 c_p^i}{\partial r^2} + \frac{\beta}{r} \frac{\partial c_p^i}{\partial r} - K^i \lambda^i c_p^i + K^{i-1} \lambda^{i-1} c_p^{i-1} \qquad (10)$$

$$N^i = D_p \varepsilon_p \frac{\partial c_p^i}{\partial r} \quad r=\text{interface} \qquad (11)$$

Spherical blocks of rock matrix with a radius = r_0 means:

$$\alpha = -3(1-\varepsilon_f)/\varepsilon_f r_0 \qquad (12)$$

$$\beta = 2 \qquad (13)$$

The inflow boundary condition is a concentration boundary with a concentration given by:

$$C_{i,in} = \text{Release}_{i,min} \cdot Q_{eq,min} \qquad (\text{Bq/l}) \qquad (14)$$

Where min stands for the limiting part of the near field model (Dissolution rate of the fuel matrix, solubility in an oxidizing environment or solubility in a reducing environment).

The outflow boundary condition used is

$$c \to 0 \text{ for } z \to \infty \qquad (15)$$

The flux at the end of the migration distance is calculated as

$$\Phi_{i,out} = Q_{eq}\left[C_{i,out} - \frac{D_L \cdot \varepsilon_f}{U_0} \cdot \left.\frac{\partial C_i}{\partial x}\right|_{out}\right] \qquad (16)$$

The calculations for single nuclides with a decaying band release as source term have in most of the cases been made with the analytical code NUCDIF (Rasmuson A,neretnieks I, 1982)

All the other calculations have been made with the TRUCHN code (Rasmuson et. al., 1982) . As this code in its present form is limited to maximum three nuclides of a decay chain and the source term description has to be either constant concentration for one nuclide or decaying band release for up to three members of a nuclide chain an approximation of the more detailed source term model outlined in 2.2.2 has to be utilized.

It can for instance be shown that the release to the biosphere of shortlived daughter nuclides with reasonably high retention will solely consist of the material formed during the migration of its parent nuclide and the primary release of the daughter nuclide will be insignificant for the migration calculation. One further approximation that has to be used is for the case with vastly differing decay constants for the different members of a decay chain. This in many cases forces the TRUCHN model to utilize so short timesteps that the calculation for the whole decay chain can not be followed all the way to the maximum release without an excessive cost in terms of computer resources. The results from the test calculations that have been made usually show that for the case with successively shorter halflives when descending the decay chain means that release of a daughter nuclide assymptotically approaches the value predicted by the reconcentration concept (Rasmuson et.al., 1982). This means that the ratio of activity release of a nuclide to that of its parent nuclide is inversely proportional to the ratio of their K_d-values provided that the migration distance in terms of halflives for the daughter nuclide is sufficiently long for the "equilibrium" to be established and that the halflife of the daughter nuclide is much shorter than the halflife of its parent nuclide.

Also when the release of a daughter nuclide, in grams per year, grossly exceeds the release of the parent nuclide the calculation has been divided in two steps.

i - the migration of the parent nuclide

ii - the migration of the daughter, nuclide and the rest of the decay chain originating from that nuclide.

To assess the relative importance of the primary release of radioactive material from a leaching canister for the release to the biosphere, a calculation has been made for all the nuclides as a constant concentration source term. See appendix C.

The result of this calculation is that for the parameter combinations used the primary source terms for 231Pa, 226Ra, 230Th, 229Th have small or no effect on the release to the biosphere. The release to the biosphere for these nuclides has been calculated according to the reconcentration concept.

2.3.2 Migration in colloidal form

The migration is modelled as pure transport with the velocity of the water.

The colloidal particles are assumed to be transported to the biosphere with the velocity of the flowing water. The release to the biosphere is assumed to be the same as the release to the geosphere.

3. INPUT PARAMETERS

3.1 Common parameters used

The choice of parameter values is discussed in the KBS-3 report.

- The canisters are assumed to be penetrated at 10^5 years after the closure of the repository. (10^2 years for the cases with initially defective canister).

- The respective inventories at 10^2- and 10^5 years are shown in tables 1 and 2.

- The solubilities in different media are shown in table 3.

- The K_d-values used are shown in table 4.

- The characteristic data for the rock are shown in table 5.

- The water flowrates and their corresponding equivalent flowrates are shown in table 6.

TABLE 1. Inventory at $t=10^5$ years.

Nuclide	$t_{1/2}$ (years)	Bq/canister	g/canister
Zr93	1.53E6	9.84E10	1.06E3
Tc99	2.14E5	5.70E11	9.12E2
I129	1.57E7	1.86E9	2.85E2
Sn126	1.00E5	2.38E10	2.27E1
Cs135	2.95E6	1.97E10	5.92E2
Ra226	1.60E3	6.22E10	1.70E0
Th229	7.34E3	2.54E10	3.12
Th230	8.00E4	6.22E10	8.64E1
Th232	1.41E10	9.32E4	2.30E1
Pa231	3.28E4	9.84E8	5.63E-1
U233	1.59E5	2.80E10	7.55E1
U234	2.45E5	9.84E10	4.26E2
U235	7.04E8	1.24E9	1.55E4
U236	2.34E7	1.92E10	7.71E3
U238	4.47E9	1.66E10	1.33E6
Np237	2.14E6	7.77E10	2.98E3
Pu239	2.41E4	8.81E11	3.83E2
Pu242	3.76E5	1.24E11	8.55E2

TABLE 2. Inventory at $t=10^2$ years.

Nuclide	$t_{1/2}$ (years)	Bq/canister	g/canister
H3	1.23+1	1.19+11	3.33-4
C14	5.73+3	2.86+10	1.73-1
Co60	5.27	5.18+8	1.24-5
Ni59	7.50+4	2.03+11	6.79+1
Ni63	1.00+2	1.59+13	7.57
Sr90	2.88+1	3.78+14	7.40+1
Zr93	1.53+6	1.13+11	1.21+3
Nb94	2.03+4	1.72+10	2.48
Tc99	2.14+5	7.77+11	1.24+3
I129	1.57+7	1.86+9	2.85+2
Cs135	2.95+6	2.02+10	6.08+2
Cs137	3.02+1	6.22+14	1.94+2
Sn126	1.00+5	4.71+10	4.48+1
Sm151	8.70+1	9.32+12	9.25
Eu154	8.5	2.23+11	2.21-2
Ho166m	1.20+3	2.02+8	3.04-3
Ra226	1.60+3	1.40+6	3.82-5
Th230	8.00+4	6.73+7	9.35-2
Th229	7.34+3	6.22+4	7.63-6
Th232	1.41+10	6.73+1	1.66-2
Pa231	3.28+4	2.54+6	1.45-3
U233	1.59+5	1.04+7	2.91-2
U234	2.45+5	9.32+10	4.04+2
U235	7.04+8	7.25+8	9.06+3
U236	2.34+7	1.40+10	5.84+3
U238	4.47+9	1.66+10	1.33+6
Np237	2.14+6	2.64+10	1.01+3
Pu238	8.77+1	8.81+13	1.39+2
Pu239	2.41+4	1.50+13	6.53+3
Pu240	6.57+3	1.97+13	2.35+3
Pu242	3.76+5	1.50+11	1.03+3
Am241	4.32	2.64+14	2.08+3
Am242m	1.52+2	3.94+11	1.09
Am243	7.37+3	1.71+12	2.31+2
Cm245	8.5+3	1.81+10	2.85
Cm246	4.70+3	4.56+9	3.98-1

TABLE 3. Solubility data

Substance	Solubility (g/l) Oxidizing environment	 Reducing environment
U	0.36(0.95)*	10^{-5} $(10^{-3})^{*}8\cdot10^{-2**}$
Pu	$8\cdot10^{-6}$	$3\cdot10^{-3}$
Np	0.63	$8\cdot10^{-6}$
Th	$4\cdot10^{-7}$	$4\cdot10^{-7}$
Am	$6\cdot10^{-3}$	$6\cdot10^{-3}$
Tc	High	$2\cdot10^{-7}$

* Values used for the cases with the water flowrate= 1.0 l/m^2, year and 0.3 l/m^2, year.

** Value used for some calculations on the 1.0 l/m^2,year case.

TABLE 4. Adsorption data

Substance	Mass absorption koefficient $K_d(m^3/kg)$
Co,Ni	0.2
Sr	0.04
Zr,Nb	4
Tc(ox/red)	0.0002/0.05
I	0
Cs	0.05
Ce,Nd,Eu,Ho,Sm	5
Ra	0.1
Th,Pa	5
U,Np(ox/red)	0.01/5
Pu	5 (0.5 if complex bound)
Am	5 " " " "

TABLE 5. Characteristic data for the geologic medium

- Distance between fractures = 2b = 5 m
- Water flow porosity ε_f = 10^{-4}
- Porosity of the rock matrix ε_p = 0.002
- Effective diffusivity in the rock matrix $D_p\ \varepsilon_p$ = = $5 \cdot 10^{-14}$ m^2/s
- Peclet number $U \cdot 1/D_L$ = Pe = 2 and 50
- Concentration of colloidal particles $C_{coll}=5 \cdot 10^{-4}$ kg/m^3
- Density of the rock matrix ρ_p=2700 kg/m^3

TABLE 6. Water flowrates and corresponding equivalent flowrates

U_o l/m^2, year	$Q_{eq,ox}$ (l/canister,year)	$Q_{eq,red}$ (l/canister,year)
0.03	0.32	4.5
0.1	0.57(0.79)*	15
0.3	0.88	45
1.0	1.57(2.7)*	150

* The figures in parenthesis stands for the elevated flow around an initially defective canister which begins to leach at 100 years after the fuel has been taken out from the reactor. The temperature in the immediate vicinity of the canister is assumed to be 50 degrees centigrade above the ambient temperature.

3.2 Description of the different scenarios

The four main scenarios can be summarized as in table 7.

TABLE 7. Migration scenarios used

Water flow rate U_0 (l/m^2,year)	Migration distance (m)
0.1	100
1	50
0.03	5
0.3	100

4. RESULTS

4.1 Summary of the calculations performed

The parameters describing an individual set of calculations are:

The Peclet number

Pe = 2

Pe = 50

The solubility of uranium in the reducing zone

10^{-5} g/l

10^{-3} g/l

$8 \cdot 10^{-2}$ g/l

The solubility of uranium in the oxidizing zone

0.36 g/l

0.95 g/l

Migration conditions

reducing conditions

oxidizing conditions

oxidizing conditions for uranium, reducing for the other nuclides

Canister life time

$1 \cdot 10^{5}$ years

TABLE 8 Calculations performed

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Release of dissolved material to the biosphere	53
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Release of dissolved material to the geosphere	61
Release of dissolved material to the biosphere	65
Release of colloidally bound material	69
U_0= 0.1 l/m^2,year L=100 m initially defective canister	
Release of dissolved material to the geosphere	73
Release of dissolved material to the biosphere	77
Release of colloidally bound material	81
U_0= 0.03 l/m^2,year L=5 m	
Release of dissolved material to the geosphere	85
Release of dissolved material to the biosphere	89
Release of colloidally bound material	93

4.2 Activity release to the geosphere and to the biosphere

On the following pages the release of activity to the geosphere and to the biosphere is shown.

The curves for release to the biosphere are drawn only for the time span for which the calculations were made.

For instance calculations aborted because running out of CPU-time have been included where they clearly approach a limiting maximum value.

Release of dissolved material to the geosphere.

U_o=1 l/m^2,year $C_{solub,U,ox}$= 0.95 g/l

Figure 2 The fission products except 99Tc

Figure 3 The natural decay chains and 99 Tc in oxidized form

Figure 4 The natural decay chains and 99Tc in reduced form. $C_{solub,U'red}$=8E-2 g/l

Figure 5 The natural decay chains and 99Tc in reduced form. $C_{solub,U,red}$=1E-3 g/l

TABLE 9. Existence of solid phase (years after start of leaching)

Substance	Oxidized	Reduced(8E-2g/l,U)	Reduced(1E-3g/l,U)
Tc	-	0-1E6	0-1E6
Th	0-2E8	-	5E4-1E7
Np	-	0-2E6	0-2E6
U	0-9E5	-	0-8E6
Pu	0-3E6	-	-

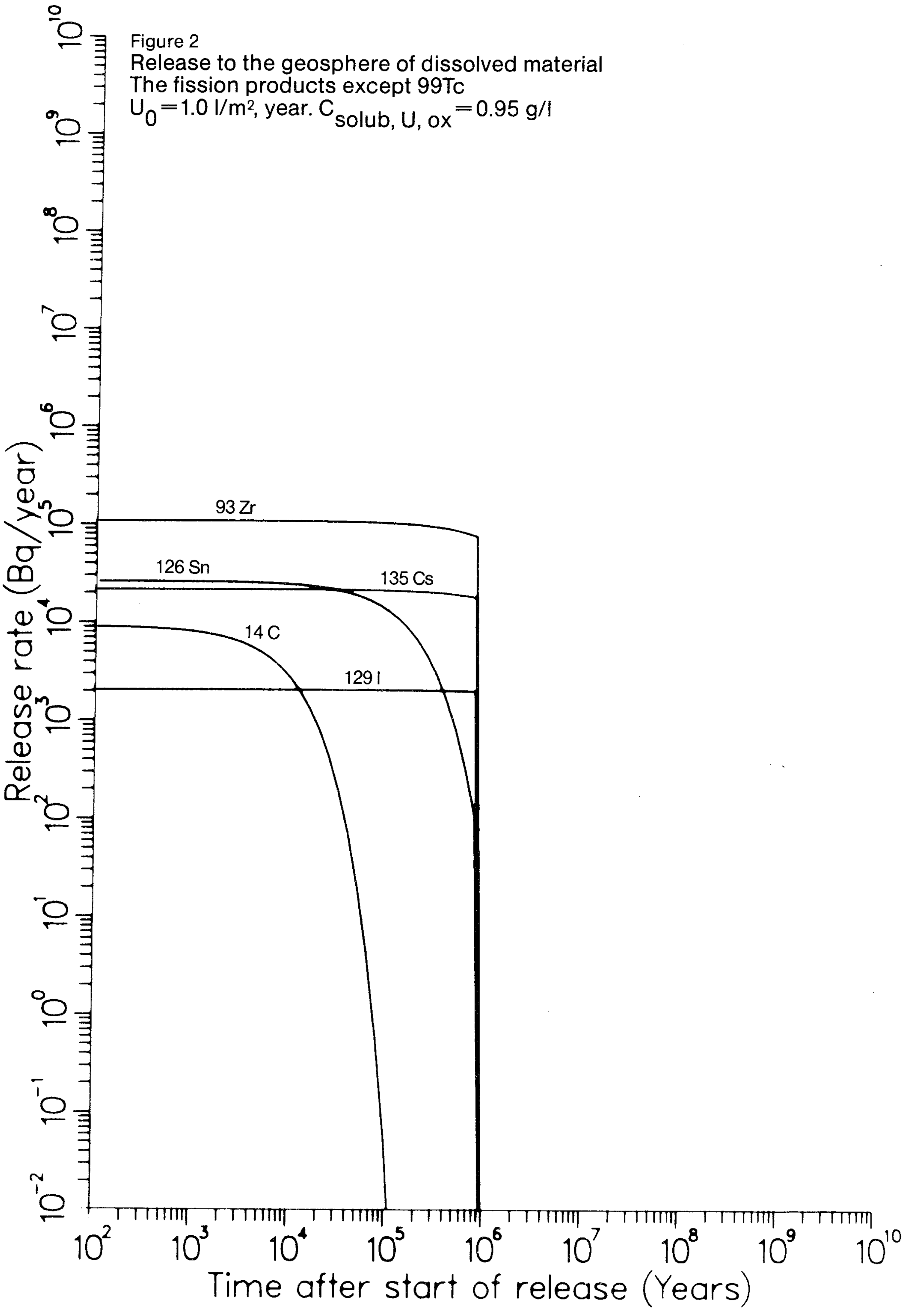


Figure 2

Release to the geosphere of dissolved material

The fission products except 99Tc

$U_0 = 1.0$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l

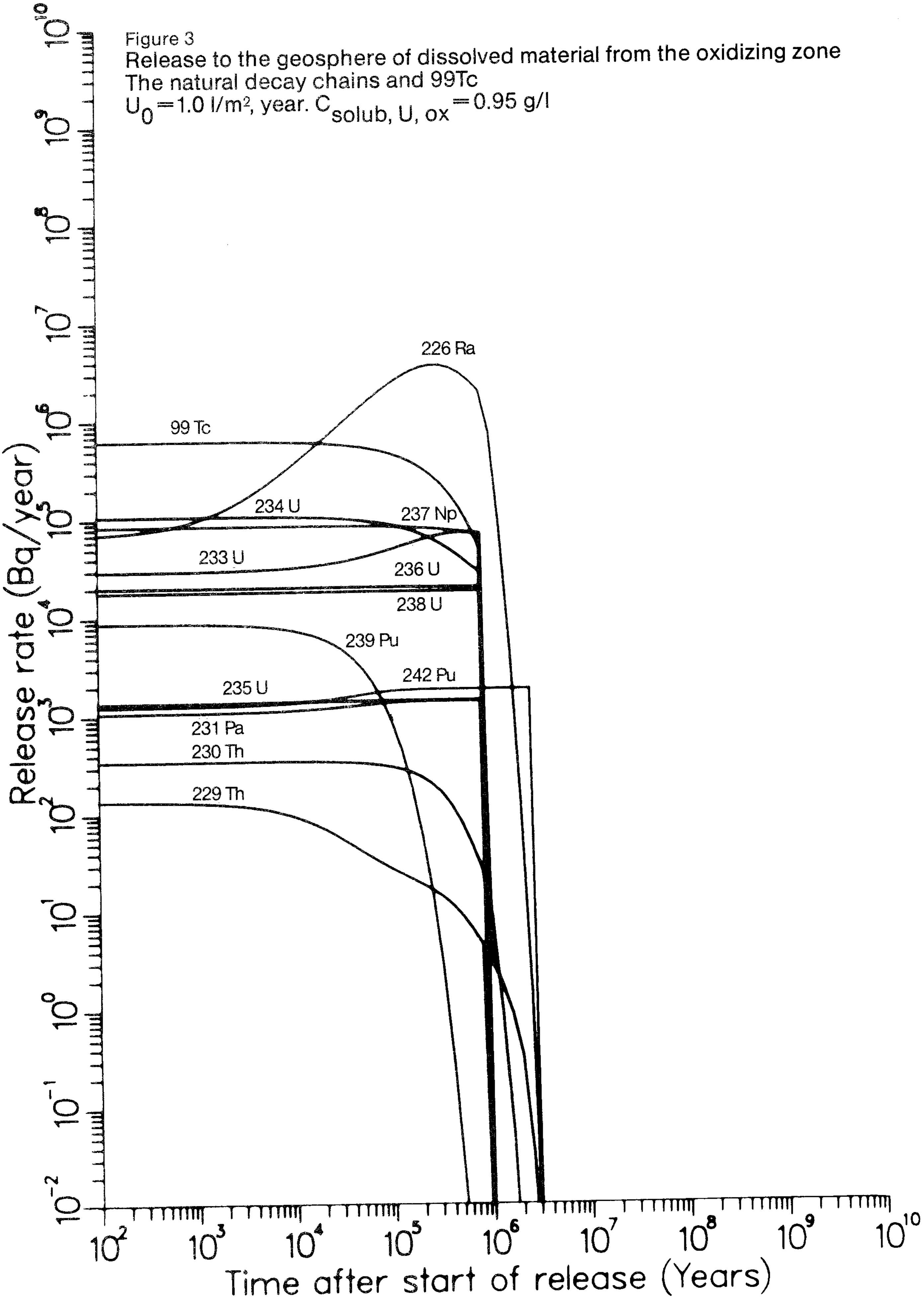


Figure 3

Release to the geosphere of dissolved material from the oxidizing zone

The natural decay chains and 99Tc

$U_0 = 1.0\ l/m^2$, year. $C_{solub,\ U,\ ox} = 0.95$ g/l

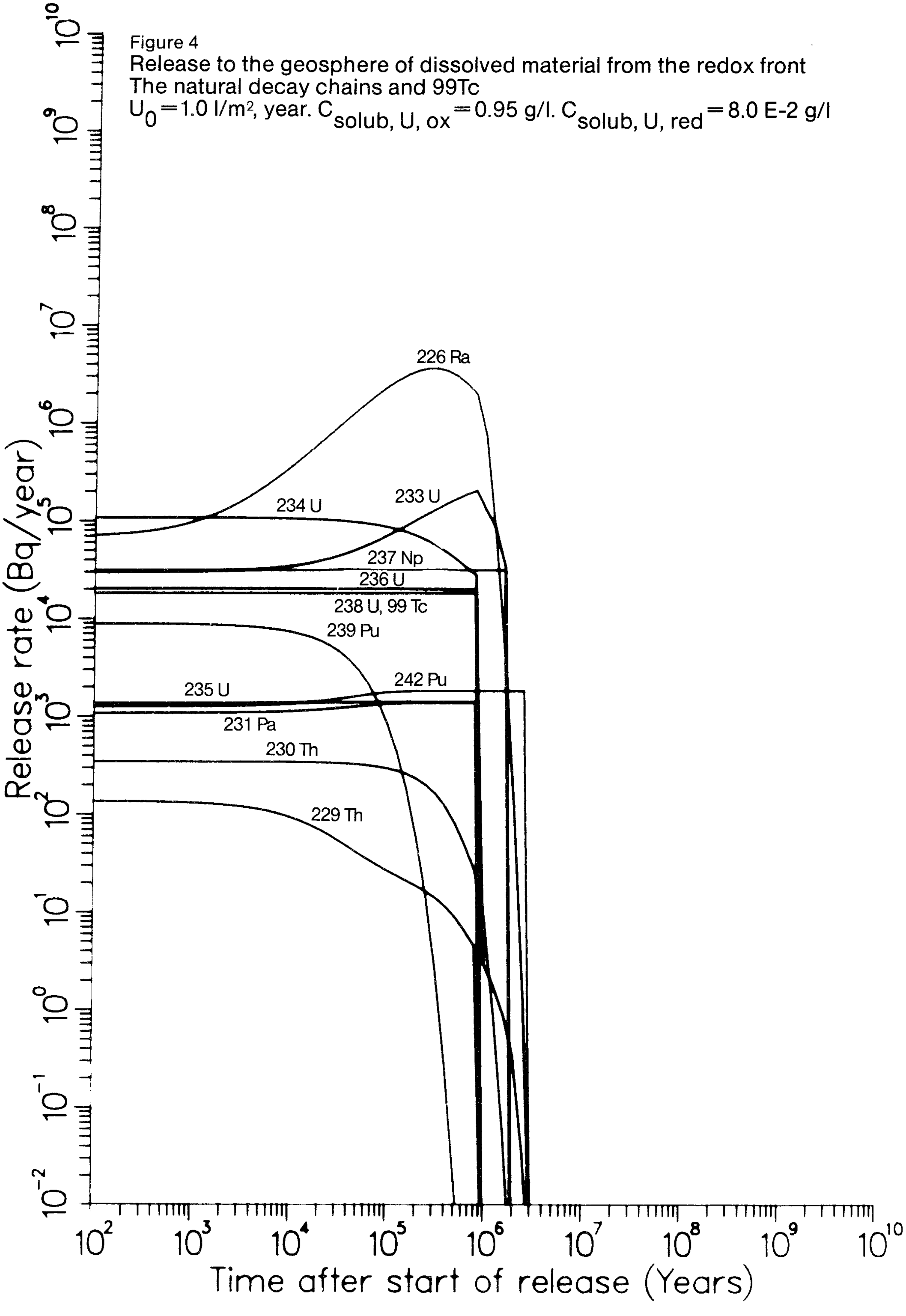


Figure 4
Release to the geosphere of dissolved material from the redox front
The natural decay chains and 99Tc
$U_0 = 1.0$ l/m², year. $C_{\text{solub, U, ox}} = 0.95$ g/l. $C_{\text{solub, U, red}} = 8.0$ E-2 g/l

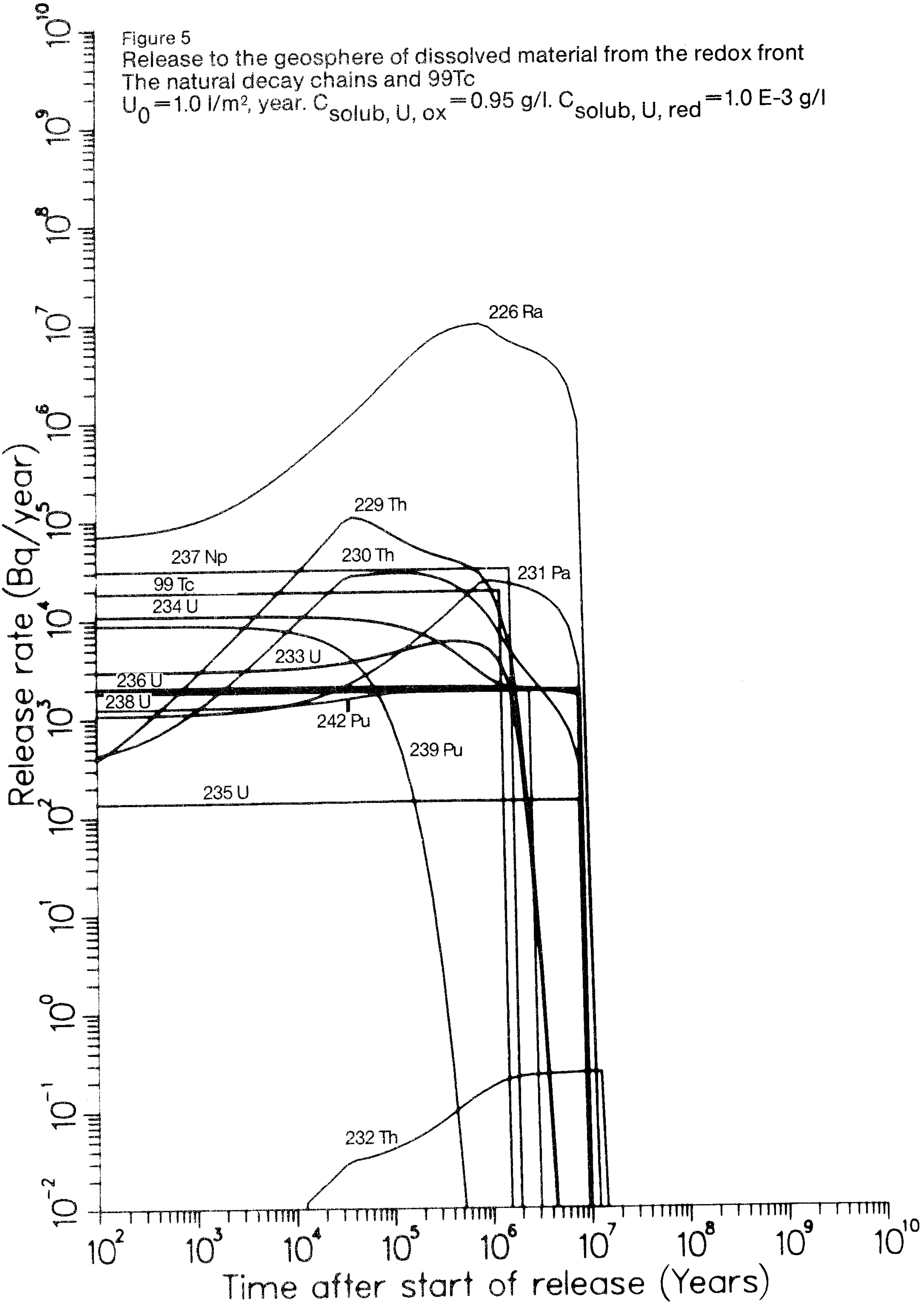


Figure 5
Release to the geosphere of dissolved material from the redox front
The natural decay chains and 99Tc
$U_0 = 1.0$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 1.0$ E-3 g/l

Release of dissolved material to the biosphere.
U_0=1 l/m^2,year $C_{solub,U,ox}$= 0.95 g/l
Migration distance = 50 m.

Figure 6	The fission product
Figure 7	The natural decay chains. Oxidizing migration conditions
Figure 8	The natural decay chains. Reducing migration conditions. $C_{solub,U,red}$=8E-2 g/l
Figure 9	The natural decay chains. Reducing migration conditions. $C_{solub,U,red}$=1E-3 g/l

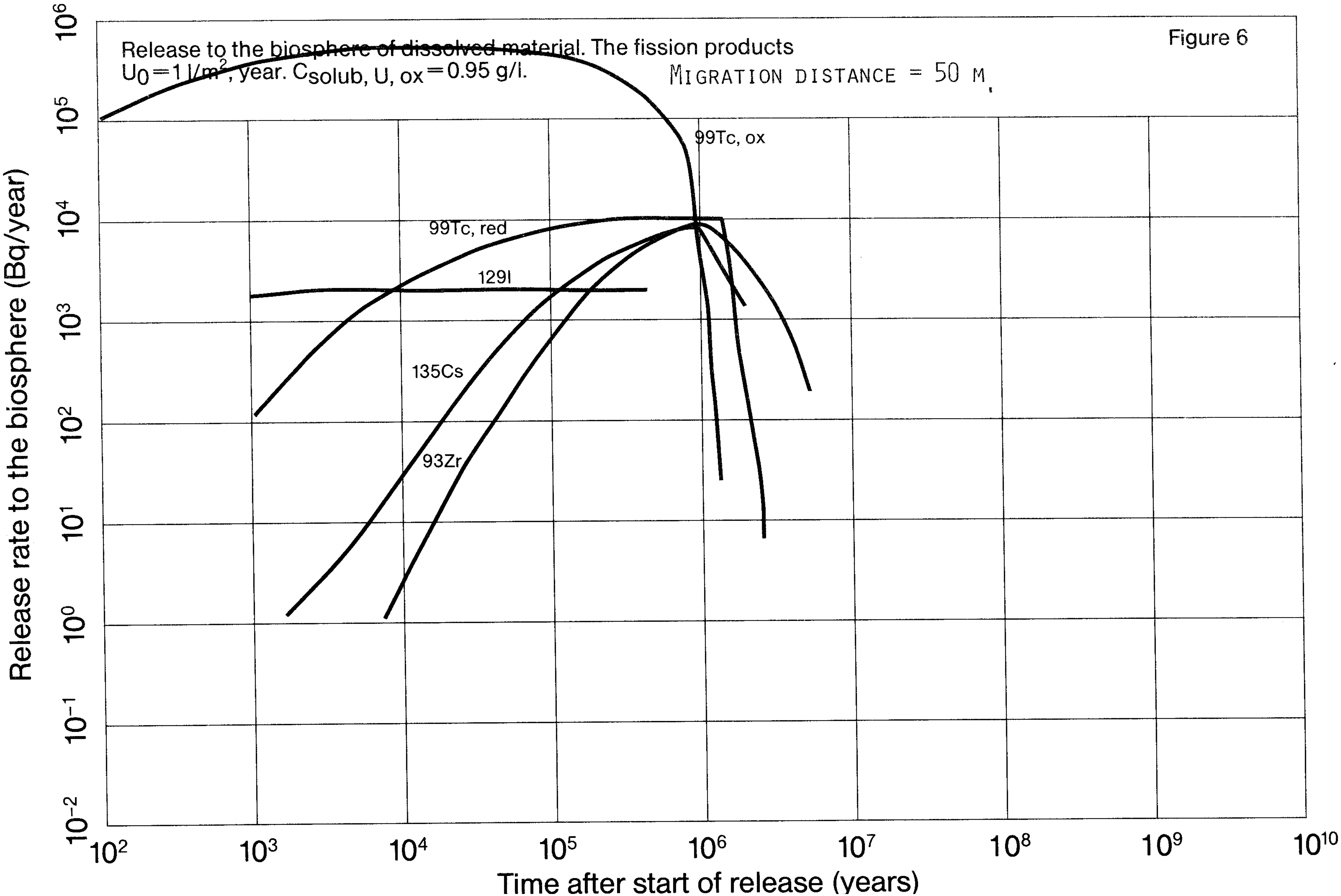


Figure 6

Release to the biosphere of dissolved material. The fission products $U_0 = 1\ l/m^2$, year. $C_{solub,\ U,\ ox} = 0.95\ g/l$.

MIGRATION DISTANCE = 50 M.

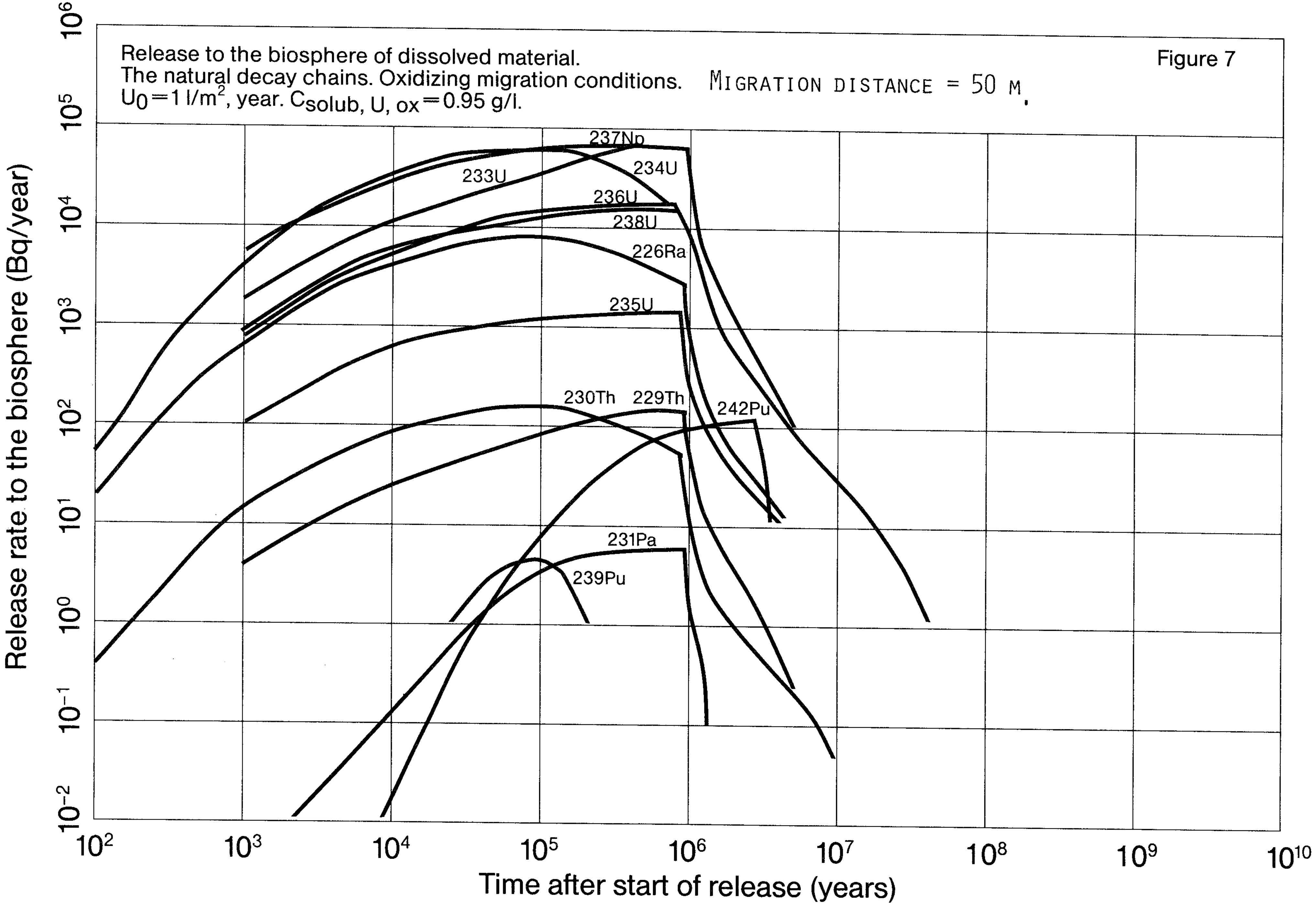

Release to the biosphere of dissolved material.
The natural decay chains. Oxidizing migration conditions. MIGRATION DISTANCE = 50 M.
$U_0 = 1$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l.

Figure 7

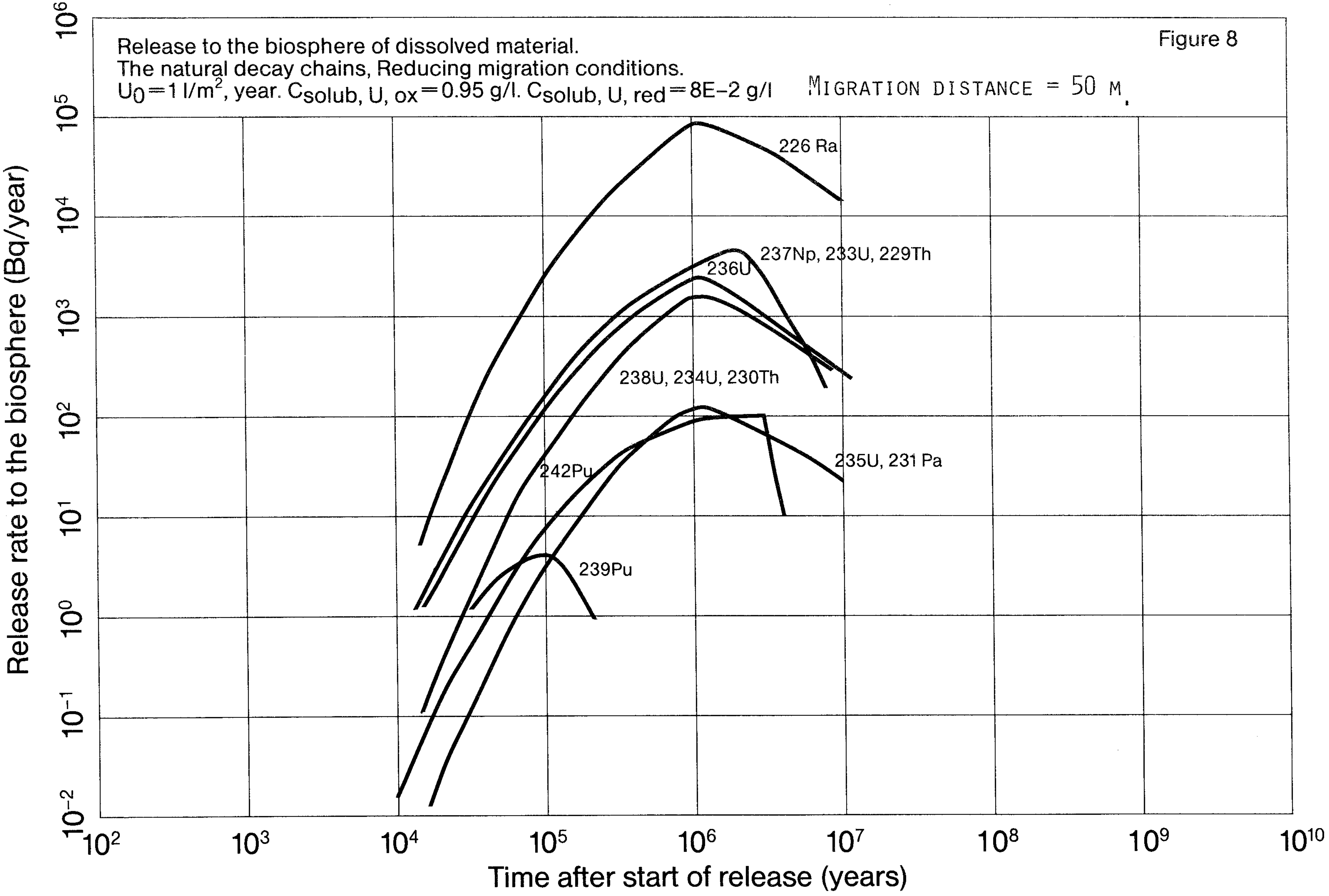


Figure 8

Release to the biosphere of dissolved material.
The natural decay chains, Reducing migration conditions.
$U_0 = 1$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 8E-2$ g/l MIGRATION DISTANCE = 50 M.

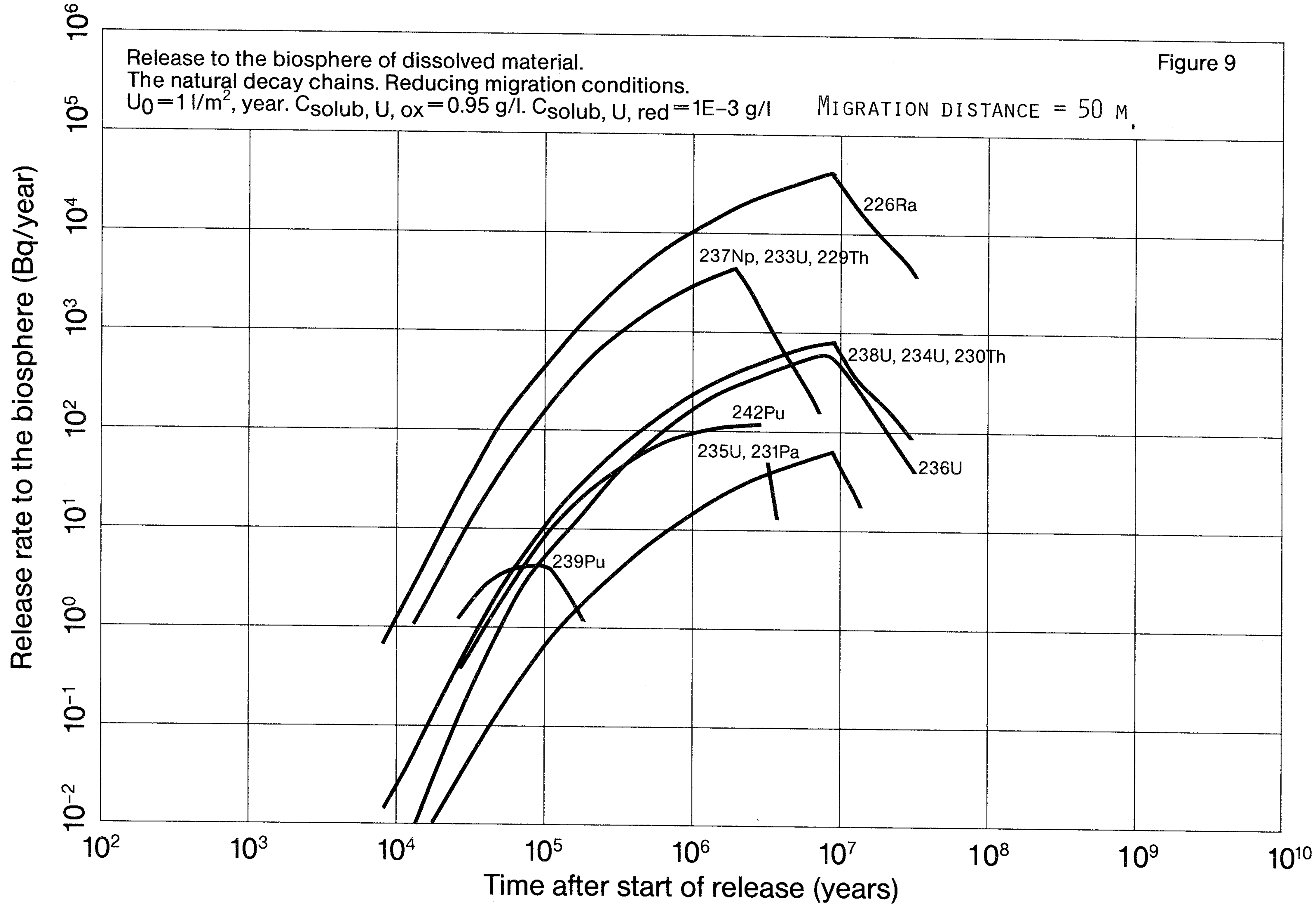


Figure 9

Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions.
$U_0 = 1$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 1E{-}3$ g/l MIGRATION DISTANCE = 50 M.

Release of colloidally bound material
U_0= 1 l/m^2,year $C_{solub,U,ox}$= 0.95 g/l

Figure 10 The fission products except 99Tc

Figure 11 The natural decay chains and 99Tc in oxidized form

Figure 12 The natural decay chains and 99Tc in reduced form. $C_{solub,U,red}$ = 8E-2 g/l

Figure 13 The natural decay chains and 99Tc in reduced form. $C_{solub,U,red}$= 1E-3 g/l

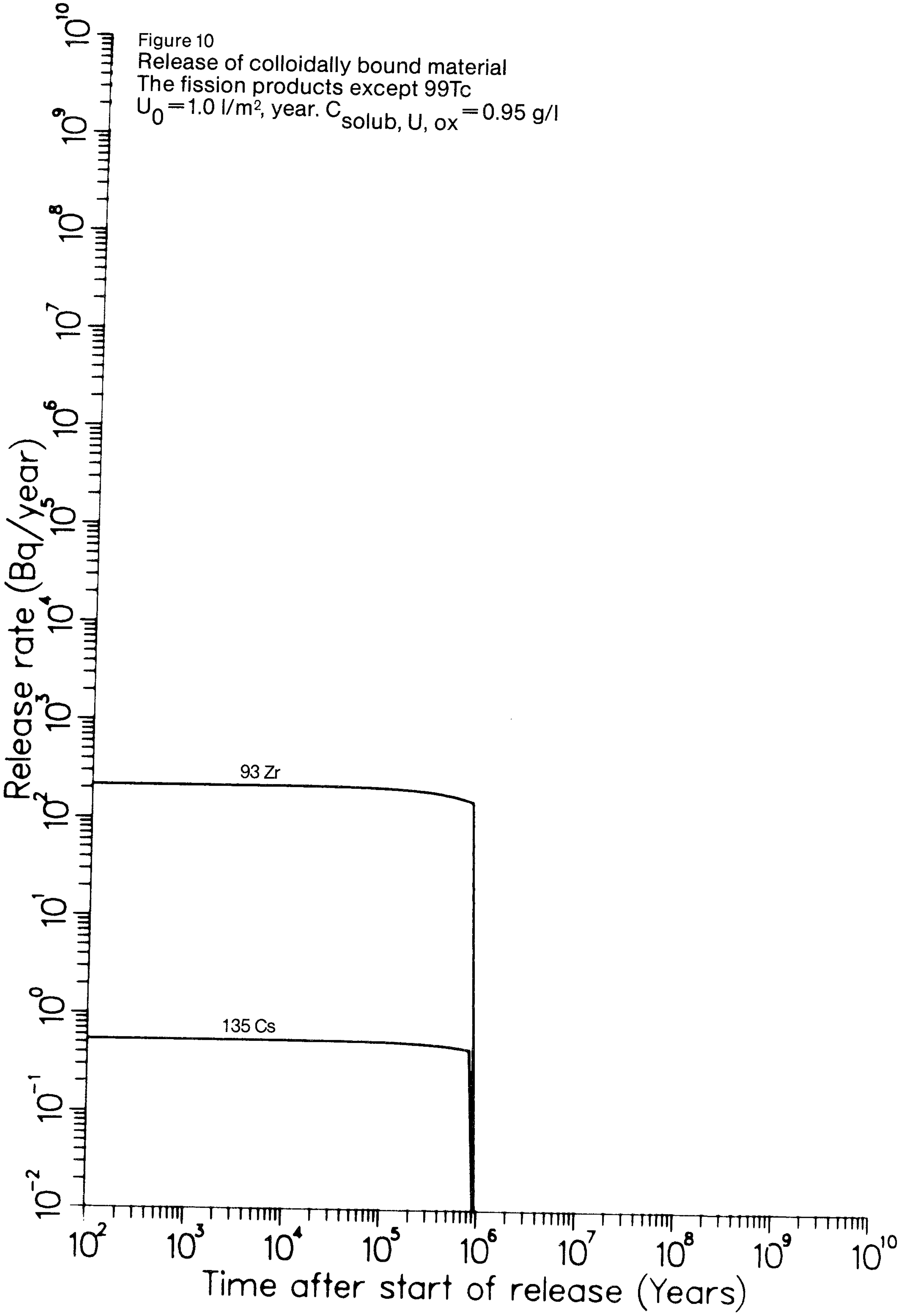


Figure 10
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 1.0\ \mathrm{l/m^2}$, year. $C_{\mathrm{solub,\ U,\ ox}} = 0.95\ \mathrm{g/l}$

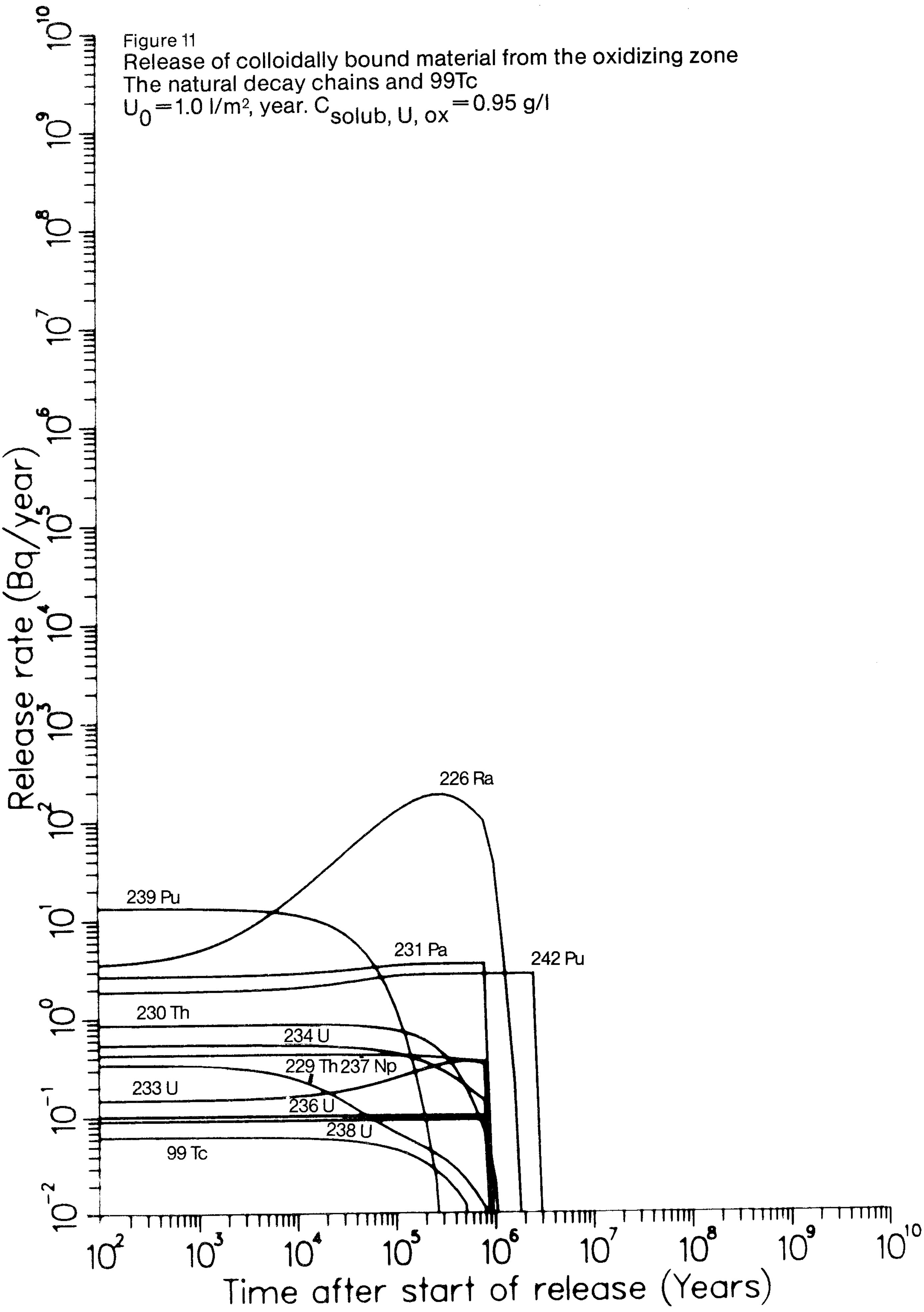


Figure 11
Release of colloidally bound material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 1.0\ l/m^2$, year. $C_{solub, U, ox} = 0.95\ g/l$

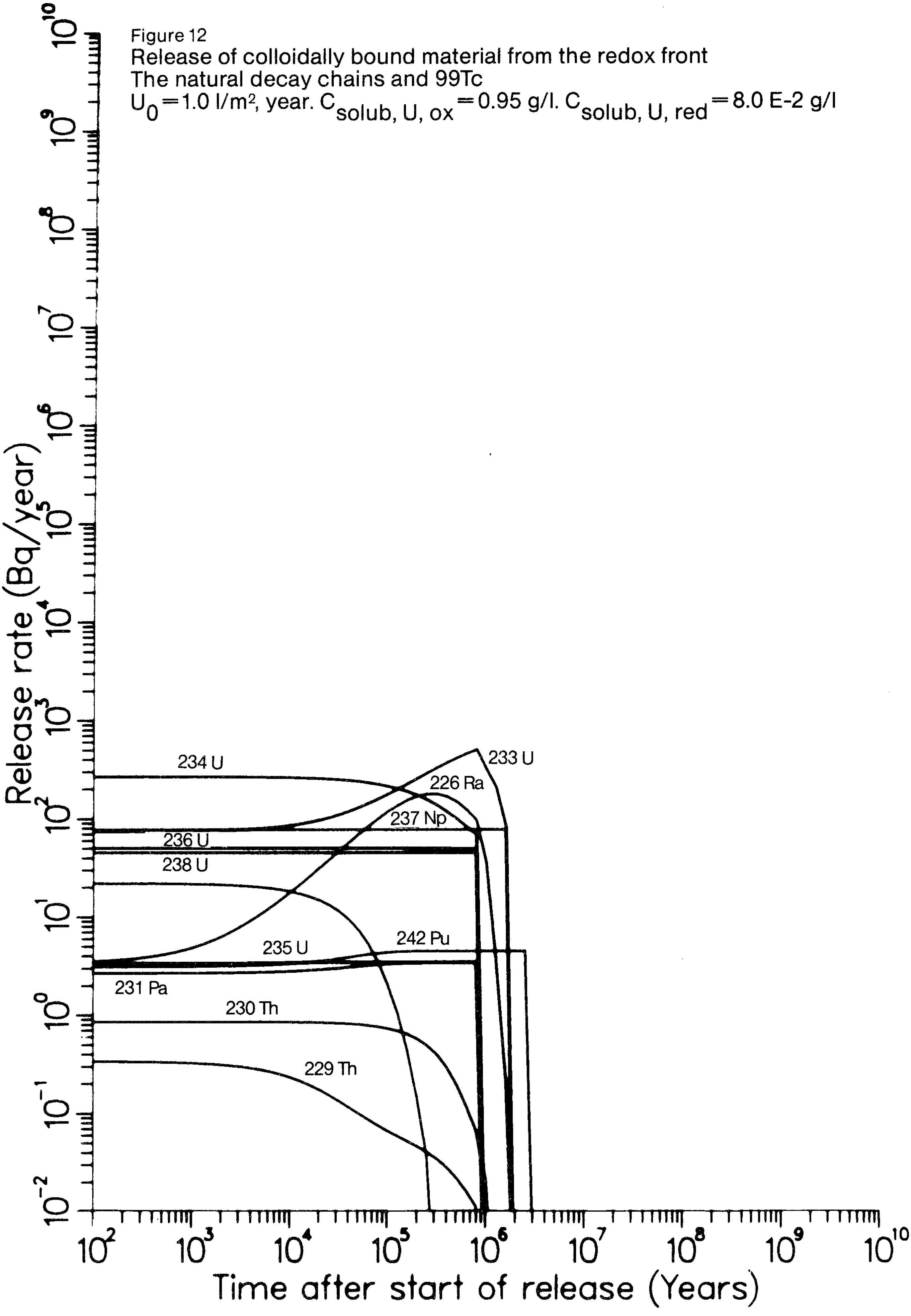


Figure 12
Release of colloidally bound material from the redox front
The natural decay chains and 99Tc
$U_0 = 1.0\ l/m^2$, year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 8.0$ E-2 g/l

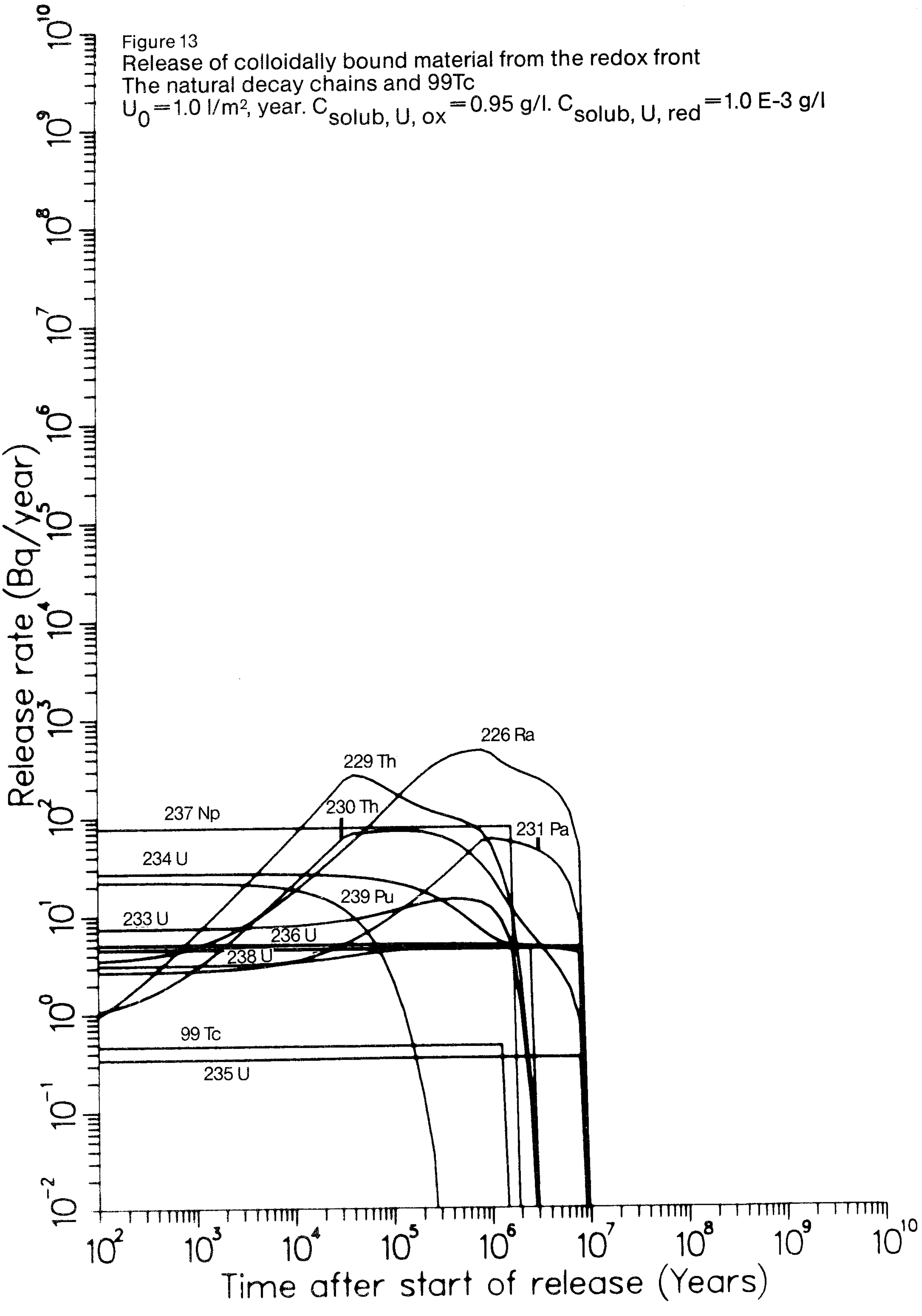


Figure 13
Release of colloidally bound material from the redox front
The natural decay chains and 99Tc
$U_0 = 1.0$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 1.0$ E-3 g/l

Comparison between Pe=2 and Pe=50.
Release of dissolved material to the biosphere.
U_o=1 l/m^2,year $C_{solub,U,ox}$=0.95 g/l. Reducing migration conditions

Figure 14 Release of 93Zr, 99Tc and 135Cs.

Comments
The larger the retention in the geologic medium the larger the effect of a lowered dispersivity.
The comparison between 99Tc and 135Cs, which have the same retention, shows that the effect of an early arrival becomes increasingly important for nuclides with shorter halflife.

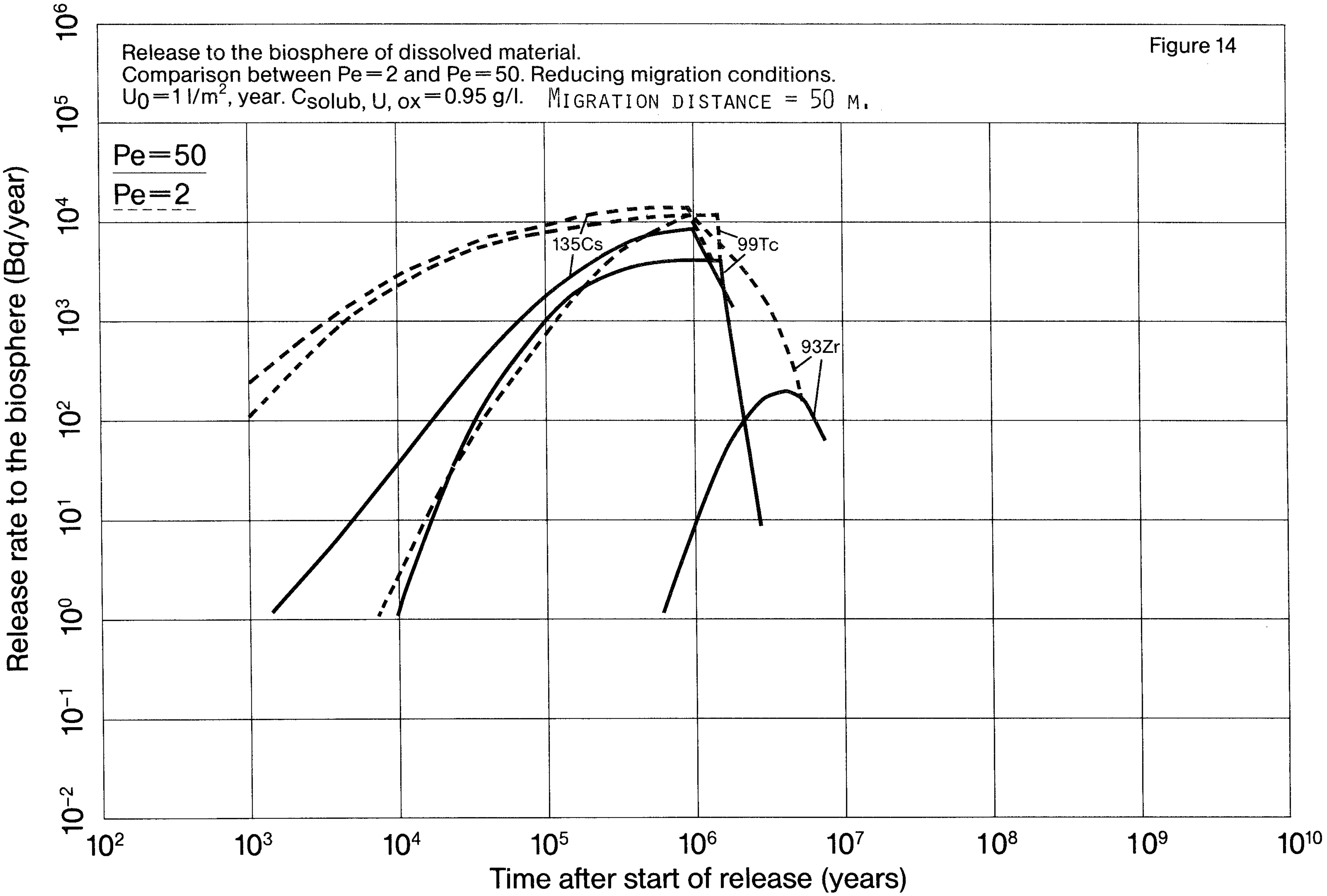


Figure 14

Release to the biosphere of dissolved material.
Comparison between Pe = 2 and Pe = 50. Reducing migration conditions.
$U_0 = 1\ \mathrm{l/m^2}$, year. $C_{\mathrm{solub,\,U,\,ox}} = 0.95$ g/l. MIGRATION DISTANCE = 50 M.

Release of dissolved material to the geosphere.
U_o=1 $1/m^2$,year $C_{solub,U,ox}$=0.95 g/l.
Initially defective canister

Figure 15	The fission products except 99Tc
Figure 16	The natural decay chains and 99Tc in oxidized form
Figure 17	The natural decay chains and 99Tc in reduced form. $C_{solub,U,red}=1\cdot 10^{-3}$ g/l.

TABLE 10. Existence times for solid phase (years after start of leaching).

Substance	The oxidizing zone	The redox front
Tc	-	0-2E6
Th	0-6E7	2E4-2E7
Np	-	0-2E6
U	0-6E5	0-1E7
Pu	0-2E6	-

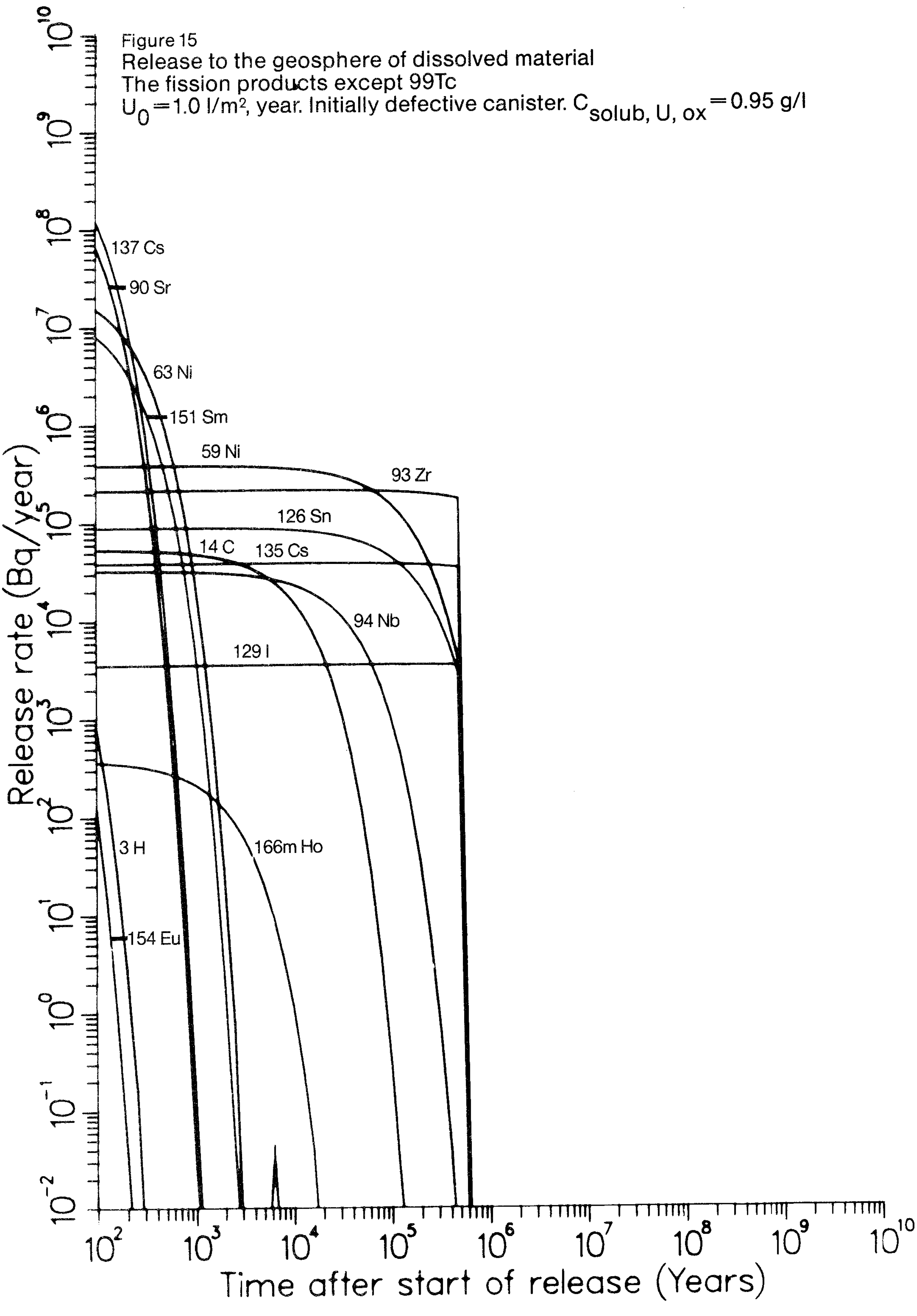


Figure 15
Release to the geosphere of dissolved material
The fission products except 99Tc
$U_0 = 1.0\ l/m^2$, year. Initially defective canister. $C_{solub, U, ox} = 0.95$ g/l

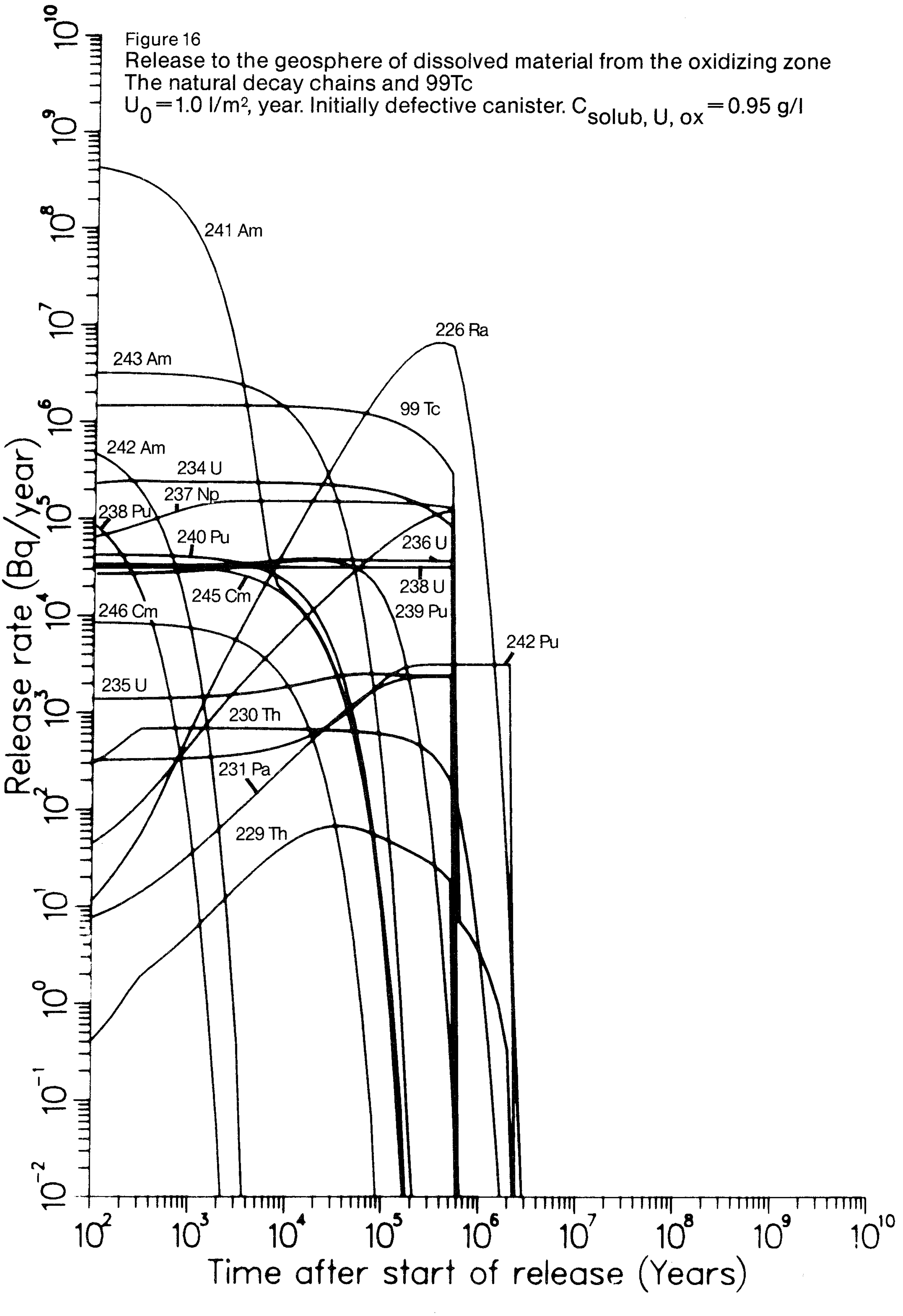


Figure 16
Release to the geosphere of dissolved material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 1.0$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.95$ g/l

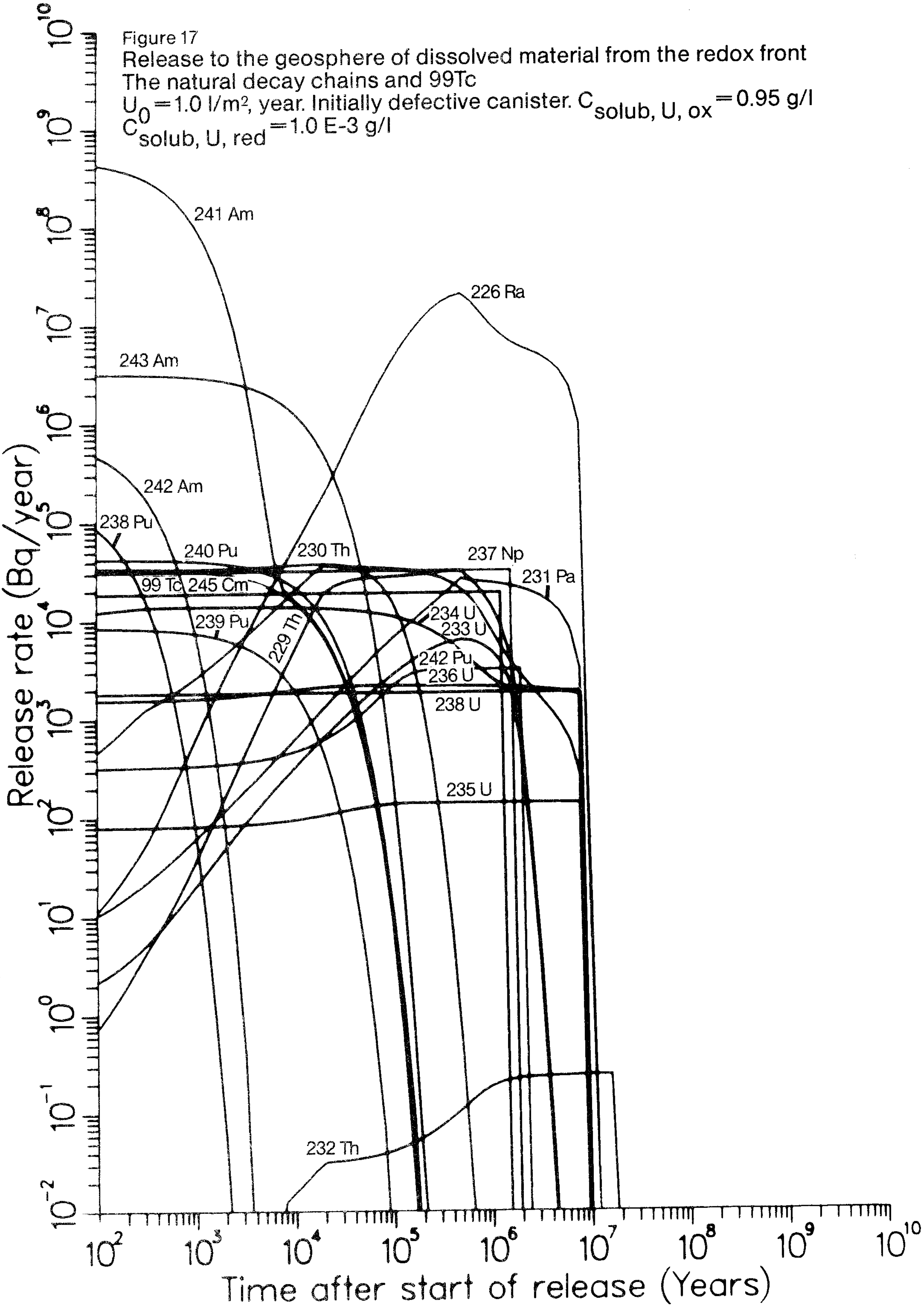


Figure 17

Release to the geosphere of dissolved material from the redox front

The natural decay chains and 99Tc

$U_0 = 1.0$ l/m², year. Initially defective canister. $C_{solub,\,U,\,ox} = 0.95$ g/l

$C^0_{solub,\,U,\,red} = 1.0$ E-3 g/l

Release of dissolved material to the biosphere.

U_o=1 l/m^2,year $C_{solub,U,ox}$=0.95 g/l.

Initially defective canister, migration distance=50 m.

Figure 18	The fission products
Figure 19	The natural decay chains. Reducing migration conditions.

Comments:

Figure 18	The curves for 14C and 126 Sn are the same as in figure 15 - release to the geosphere.

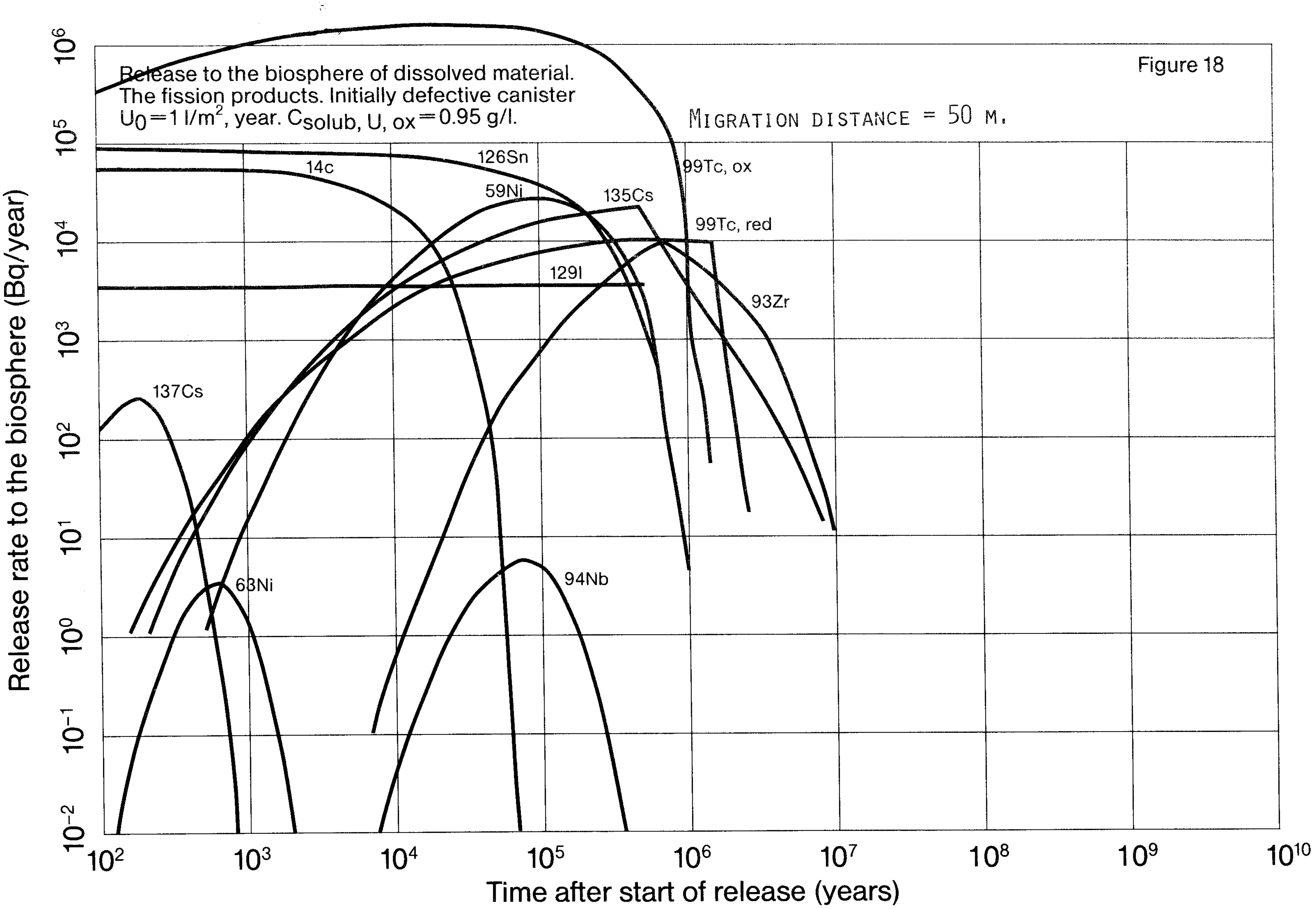


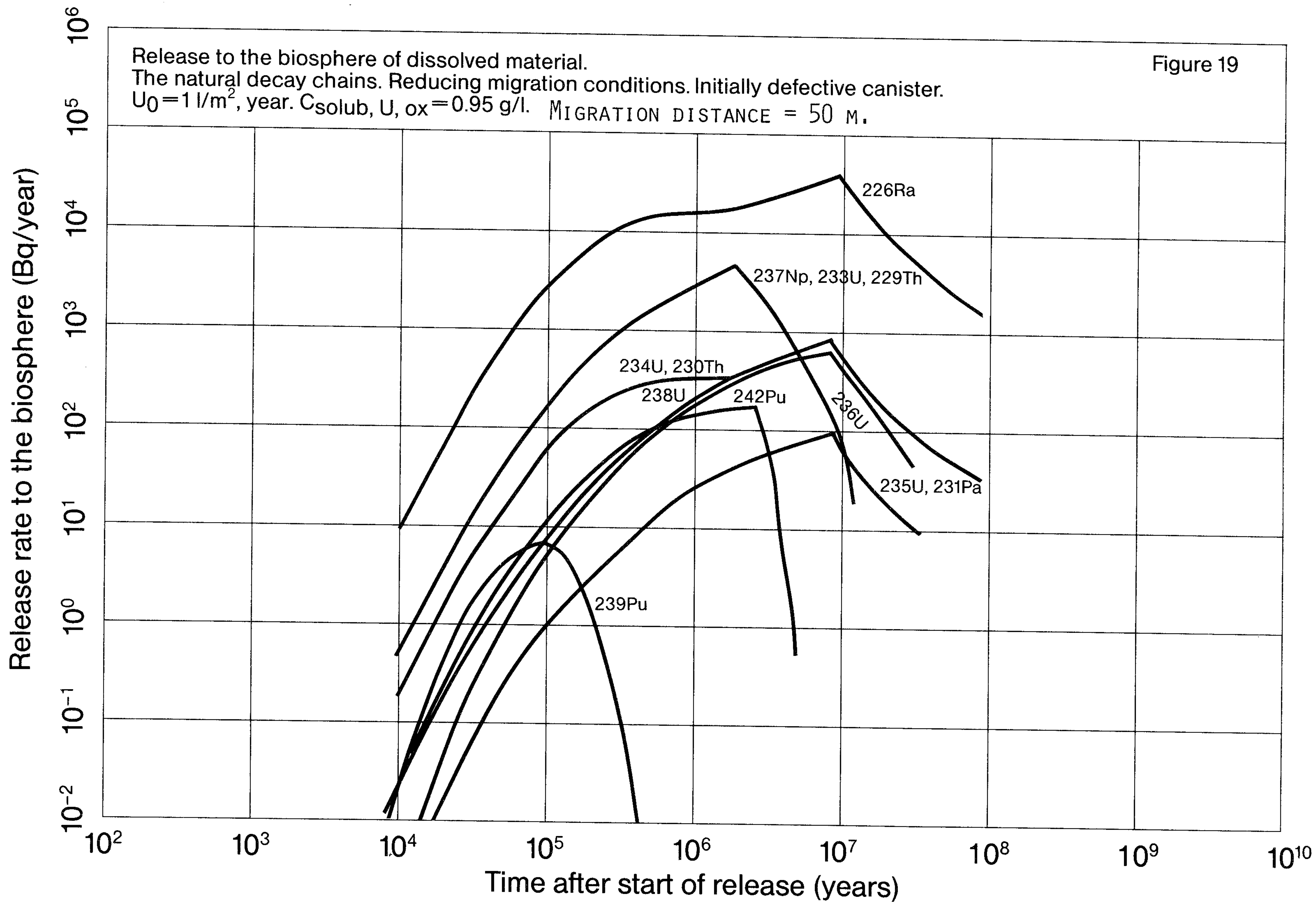
Figure 18

Release to the biosphere of dissolved material.
The fission products. Initially defective canister
$U_0 = 1$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l.

MIGRATION DISTANCE = 50 M.

Figure 19

Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions. Initially defective canister.
$U_0 = 1$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l. MIGRATION DISTANCE = 50 M.



Release of colloidally bound material.
U_o=1 l/m^2,year $C_{solub,U,ox}$=0.95 g/l.
Initially defective canister.

Figure 20	The fission products except 99Tc
Figure 21	The natural decay chains and 99Tc from the oxidizing zone
Figure 22	The natural decay chains and 99Tc from the redox front

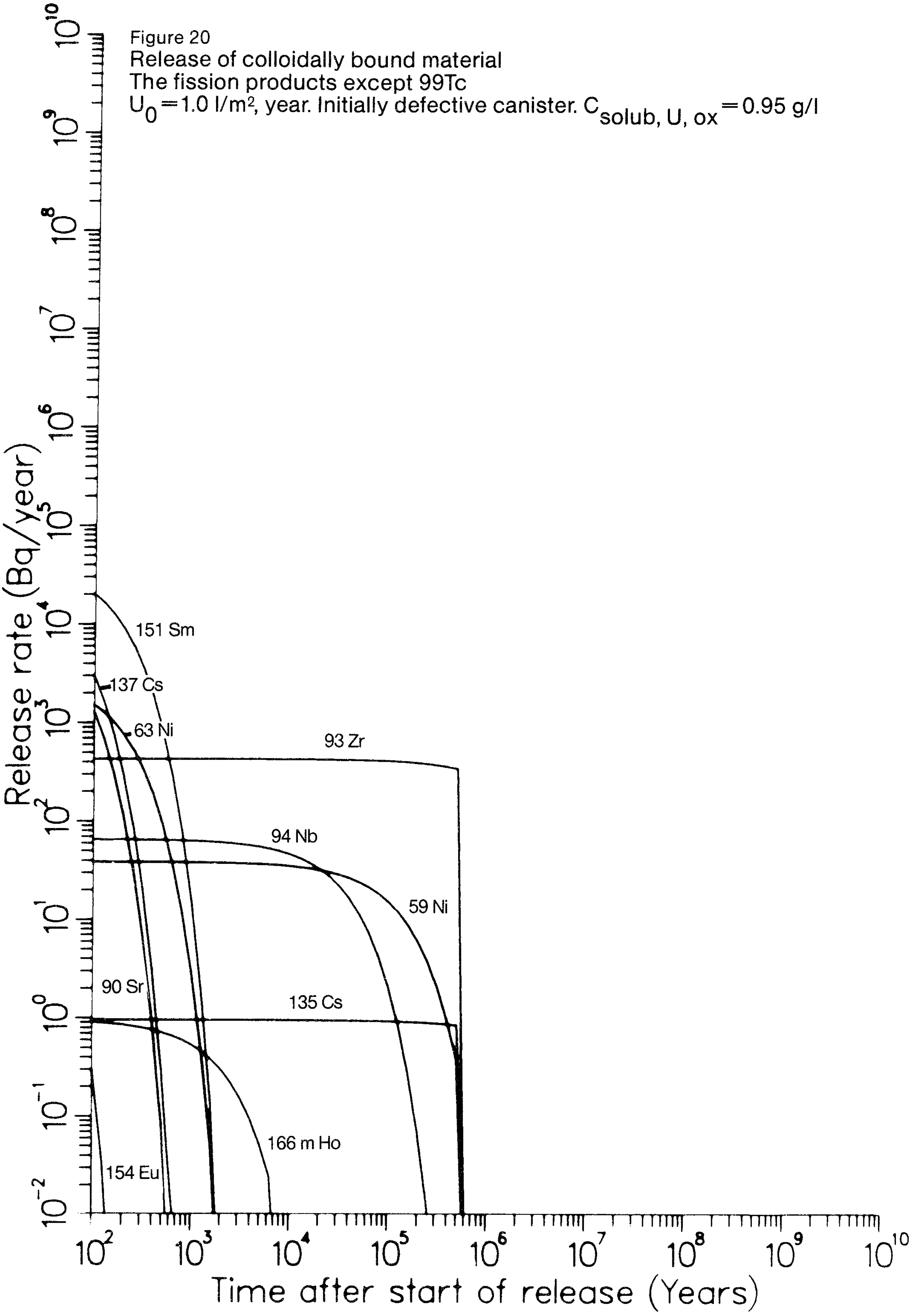


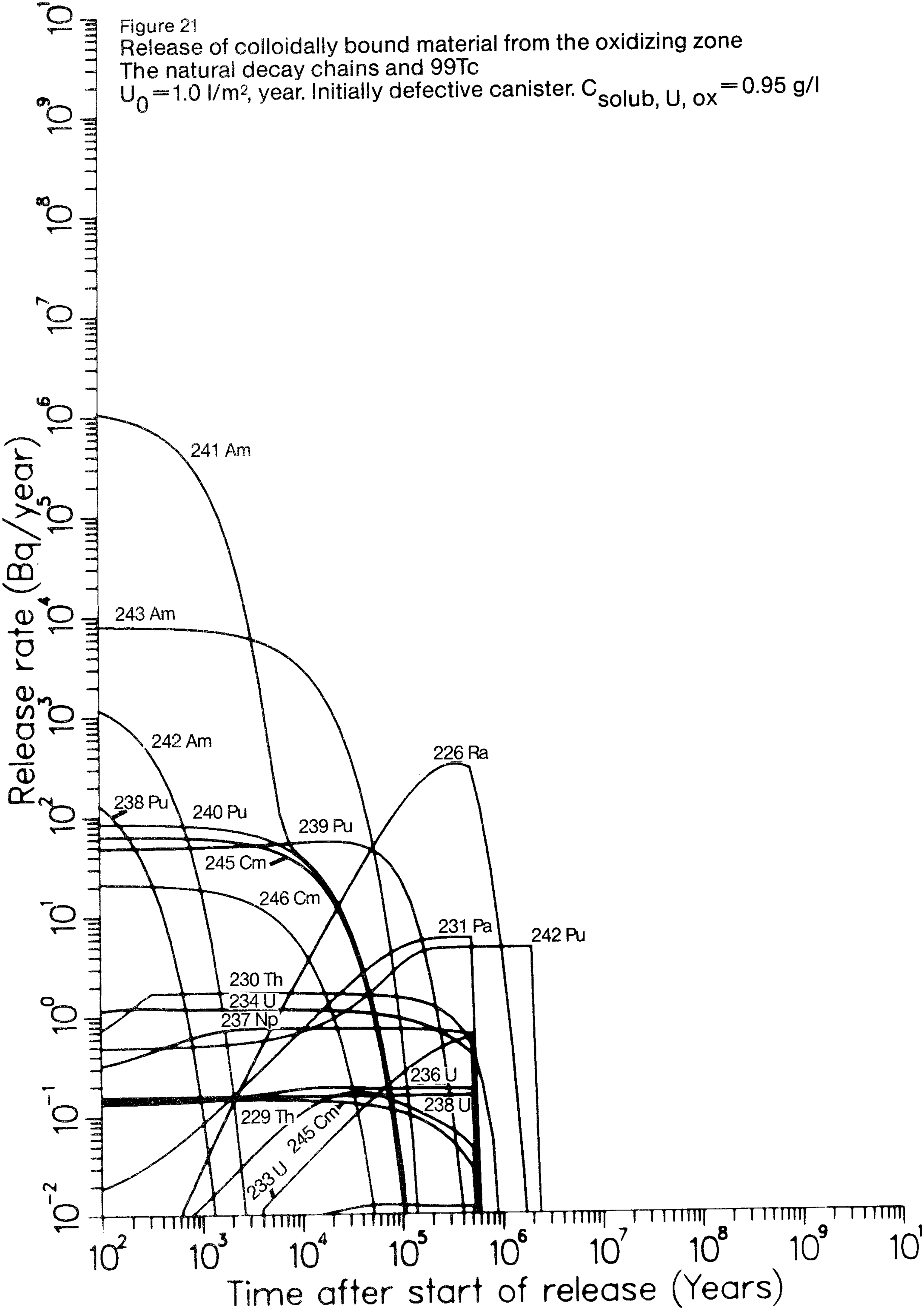
Figure 20
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 1.0\ l/m^2$, year. Initially defective canister. $C_{solub, U, ox} = 0.95$ g/l

Figure 21

Release of colloidally bound material from the oxidizing zone

The natural decay chains and 99Tc

$U_0 = 1.0$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.95$ g/l



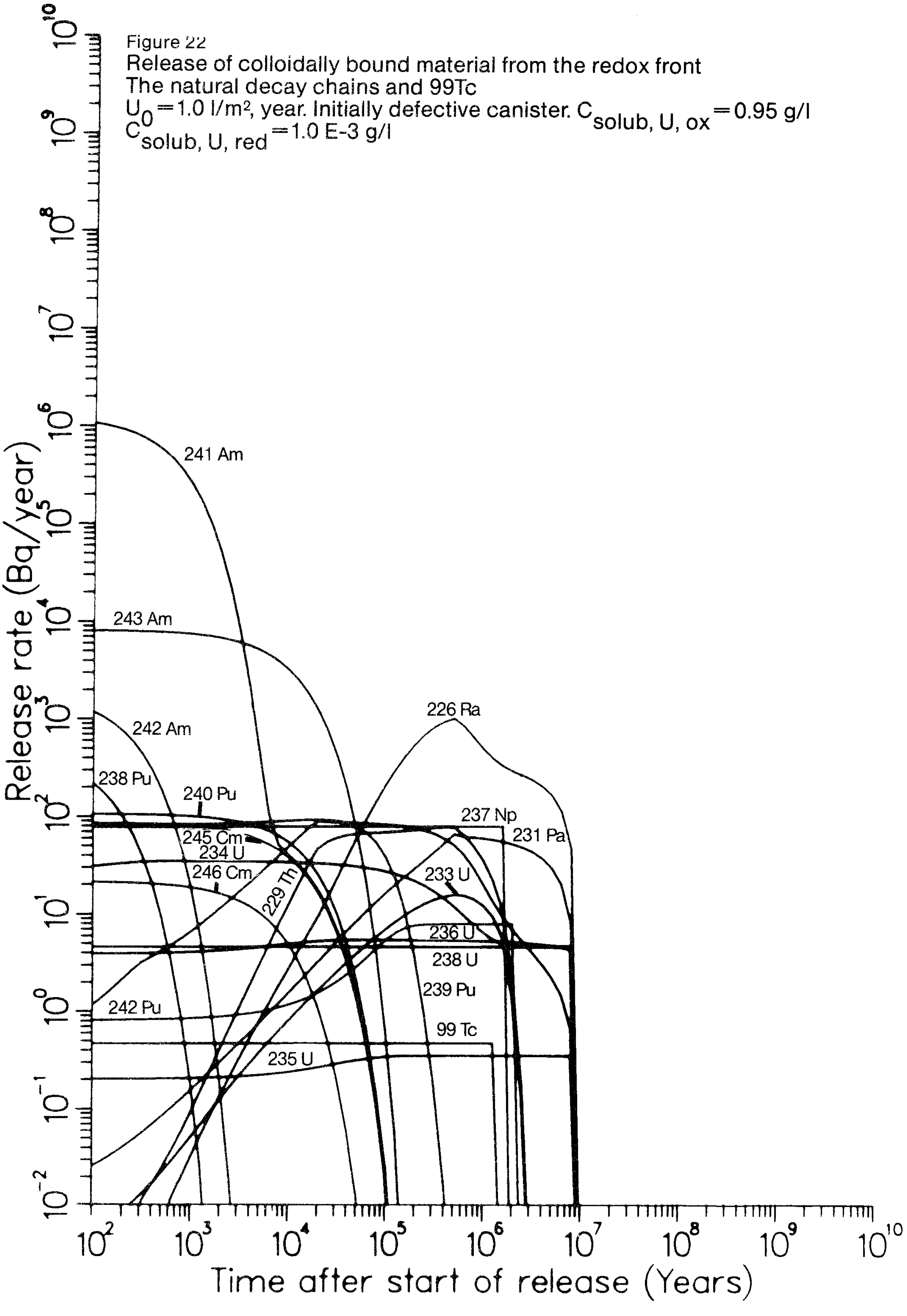


Figure 22
Release of colloidally bound material from the redox front
The natural decay chains and 99Tc
$U_0 = 1.0$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.95$ g/l
$C_{solub, U, red} = 1.0$ E-3 g/l

Release of dissolved material to the geosphere.

$U_o = 0.3\ l/m^2$,year $C_{solub,U,ox} = 0.95$ g/l.

Figure 23 The fission products except 99Tc

Figure 24 The natural decay chains and 99Tc from the oxidizing zone

Figure 25 The natural decay chains and 99Tc from the reducing zone, $C_{solub,U,red} = 8 \cdot 10^{-2}$ g/l

TABLE 11. Existence of solid phase (years after start of leaching)

Substance	The oxidizing zone	The redox front
Tc	-	0-2E6
Th	0-6E8	-
Np	-	0-5E6
U	0-2E6	
Pu	0-3E6	-

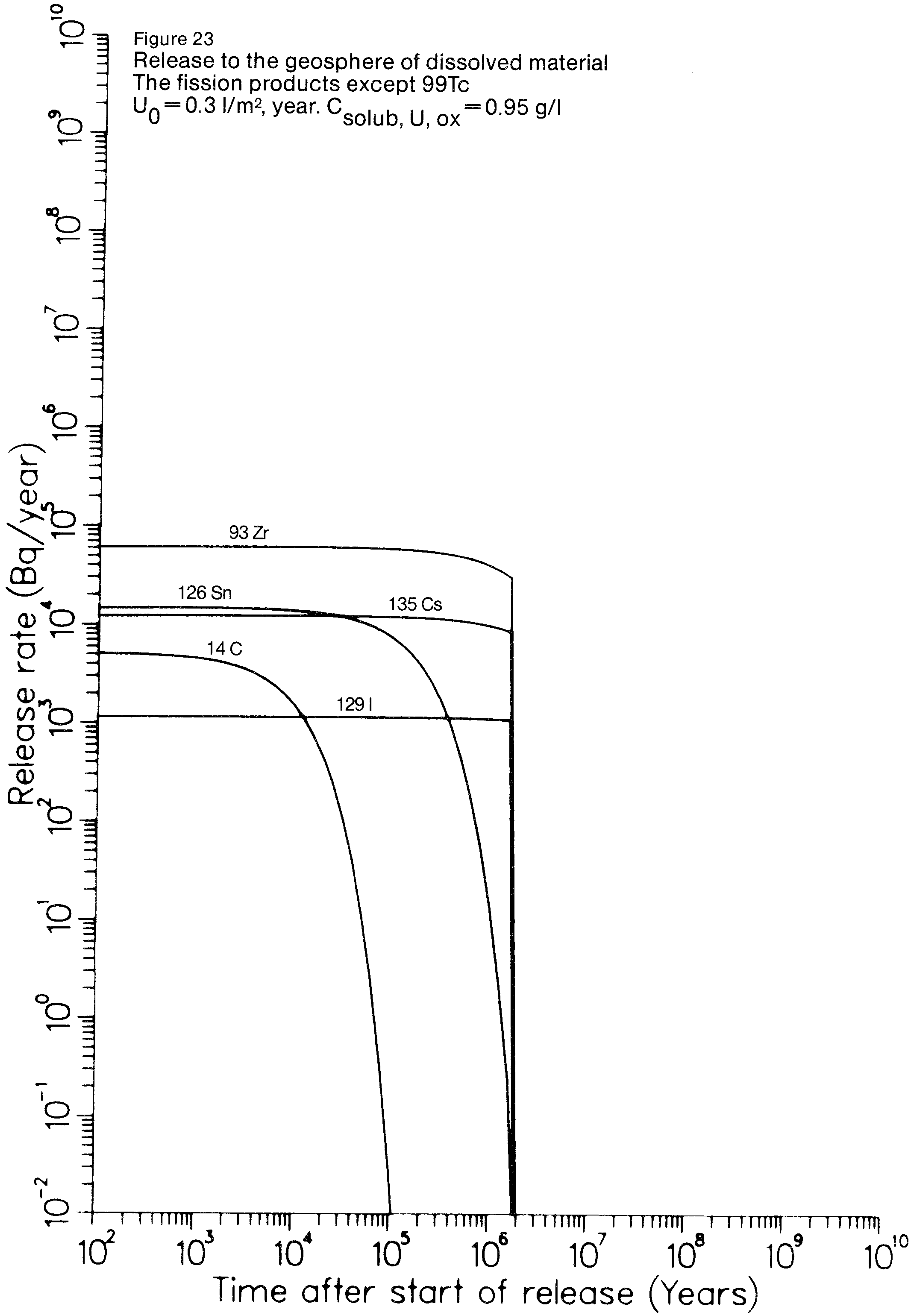


Figure 23
Release to the geosphere of dissolved material
The fission products except 99Tc
$U_0 = 0.3\ l/m^2$, year. $C_{solub, U, ox} = 0.95\ g/l$

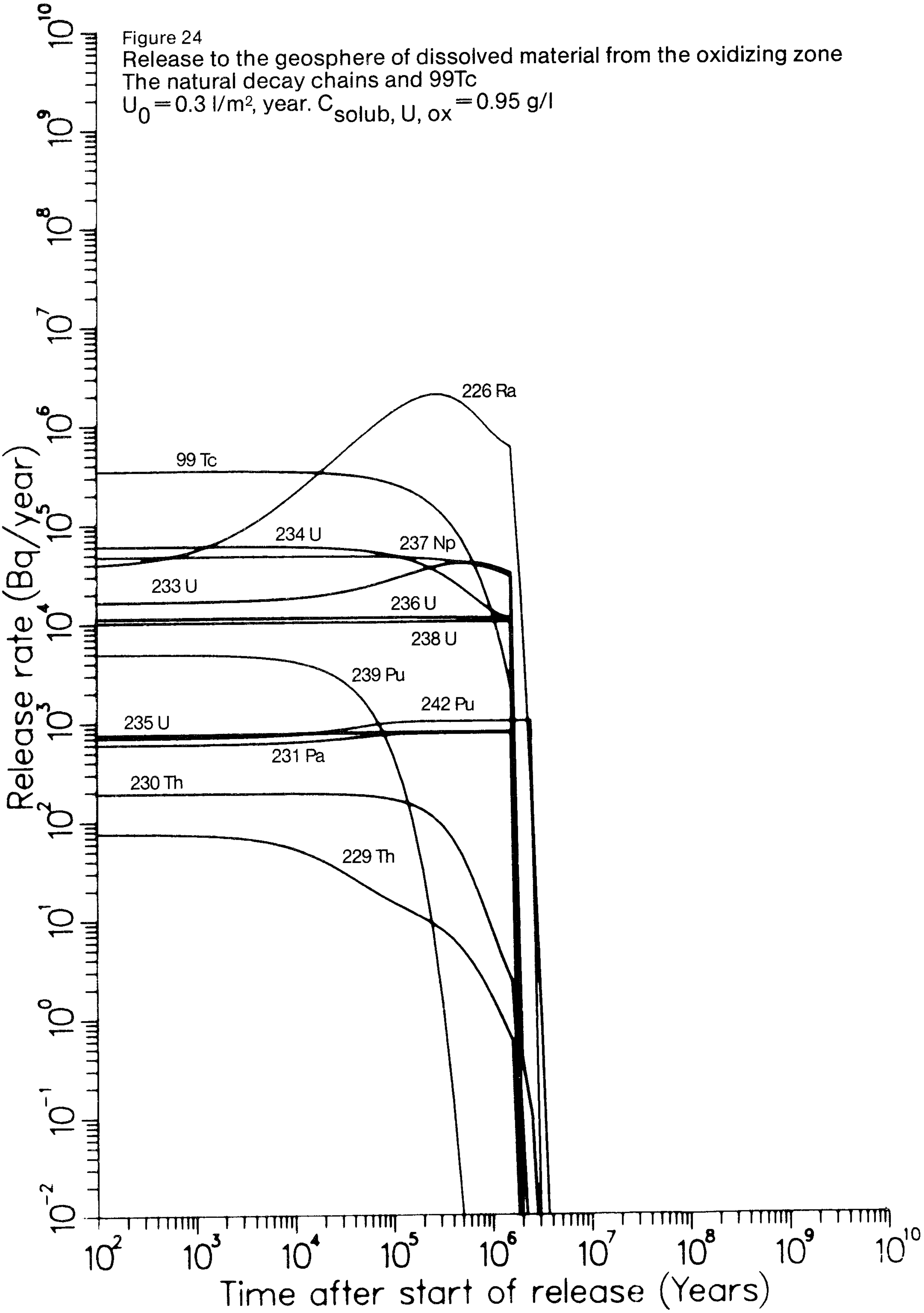


Figure 24
Release to the geosphere of dissolved material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.3$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l

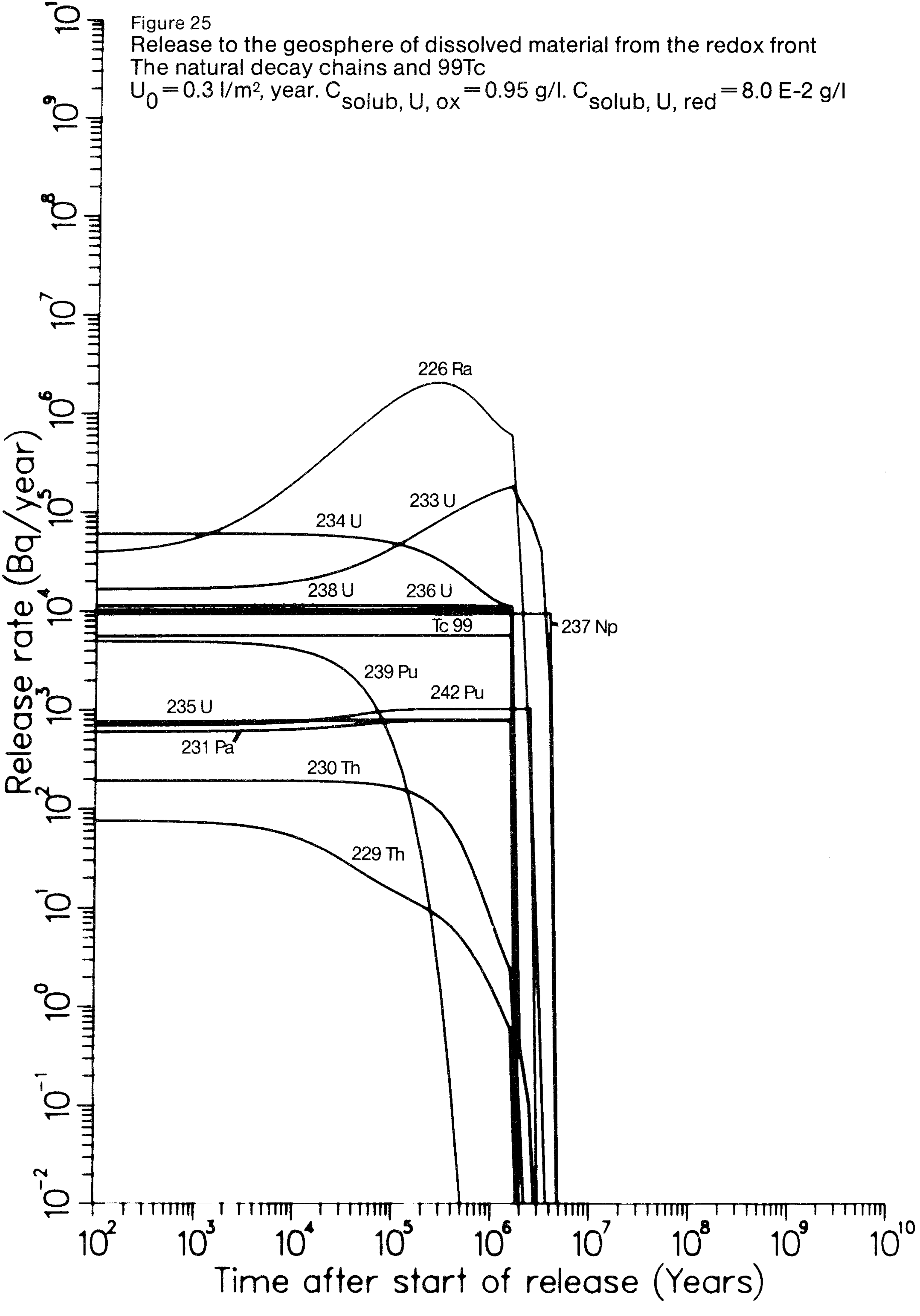


Figure 25
Release to the geosphere of dissolved material from the redox front
The natural decay chains and 99Tc
$U_0 = 0.3$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 8.0$ E-2 g/l

Release of dissolved material to the biosphere.
$U_o=0.3$ l/m^2,year $C_{solub,U,ox}=0.95$ g/l migration length = 100 m.

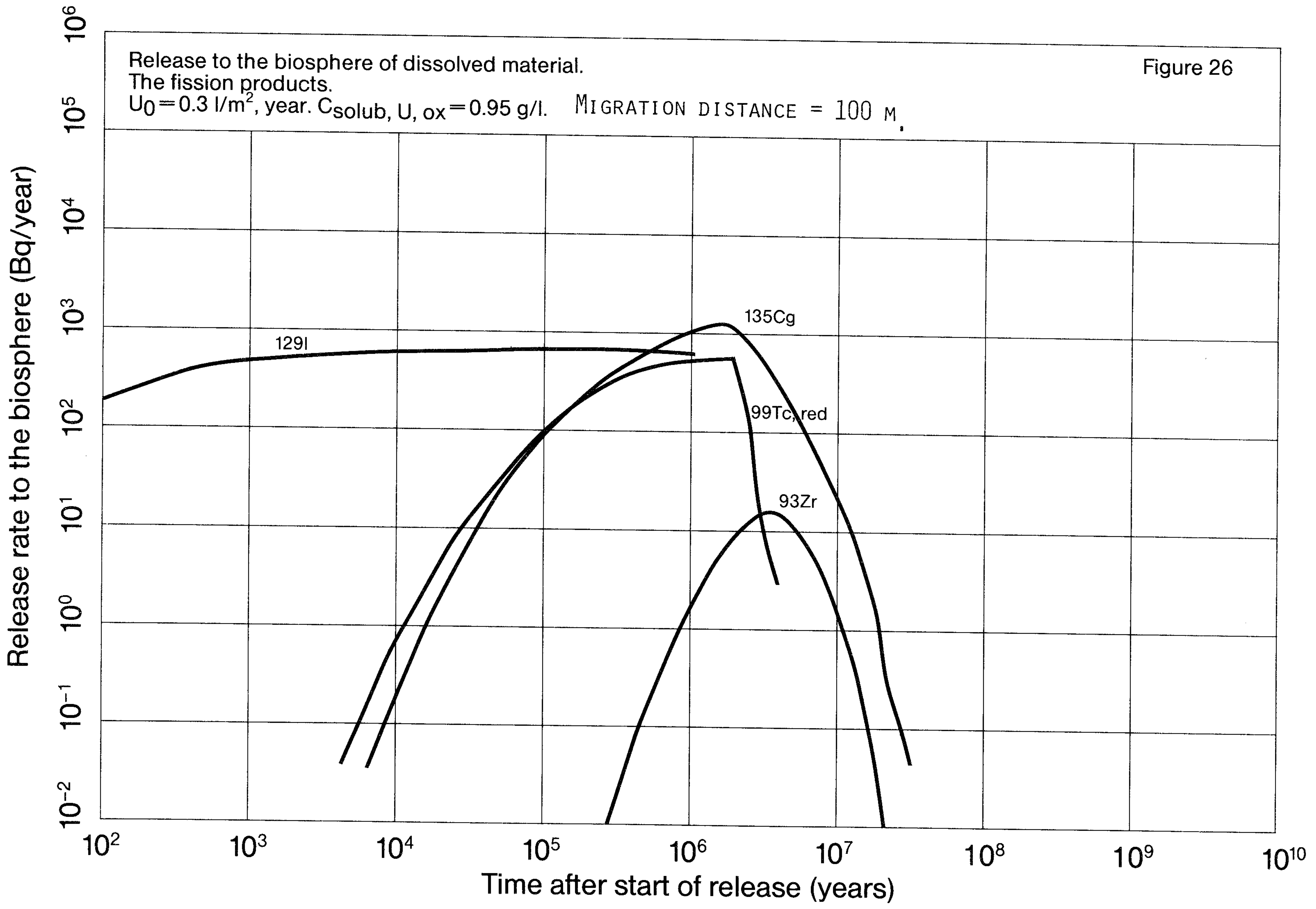
Figure 26 The fission products

Figure 27 The natural decay chains. Reducing migration conditions. $C_{solub,U,red}=8\cdot10^{-2}$ g/l

Figure 28 Oxidizing migration conditions for uranium. Reducing for the other nuclides.

Comment:

Figure 52 The curve for 233U is the sum of the curves from primary release of 233U and 233U originating from migrating 237Np.



Release to the biosphere of dissolved material.
The fission products.
$U_0 = 0.3$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. MIGRATION DISTANCE = 100 M.

Figure 26

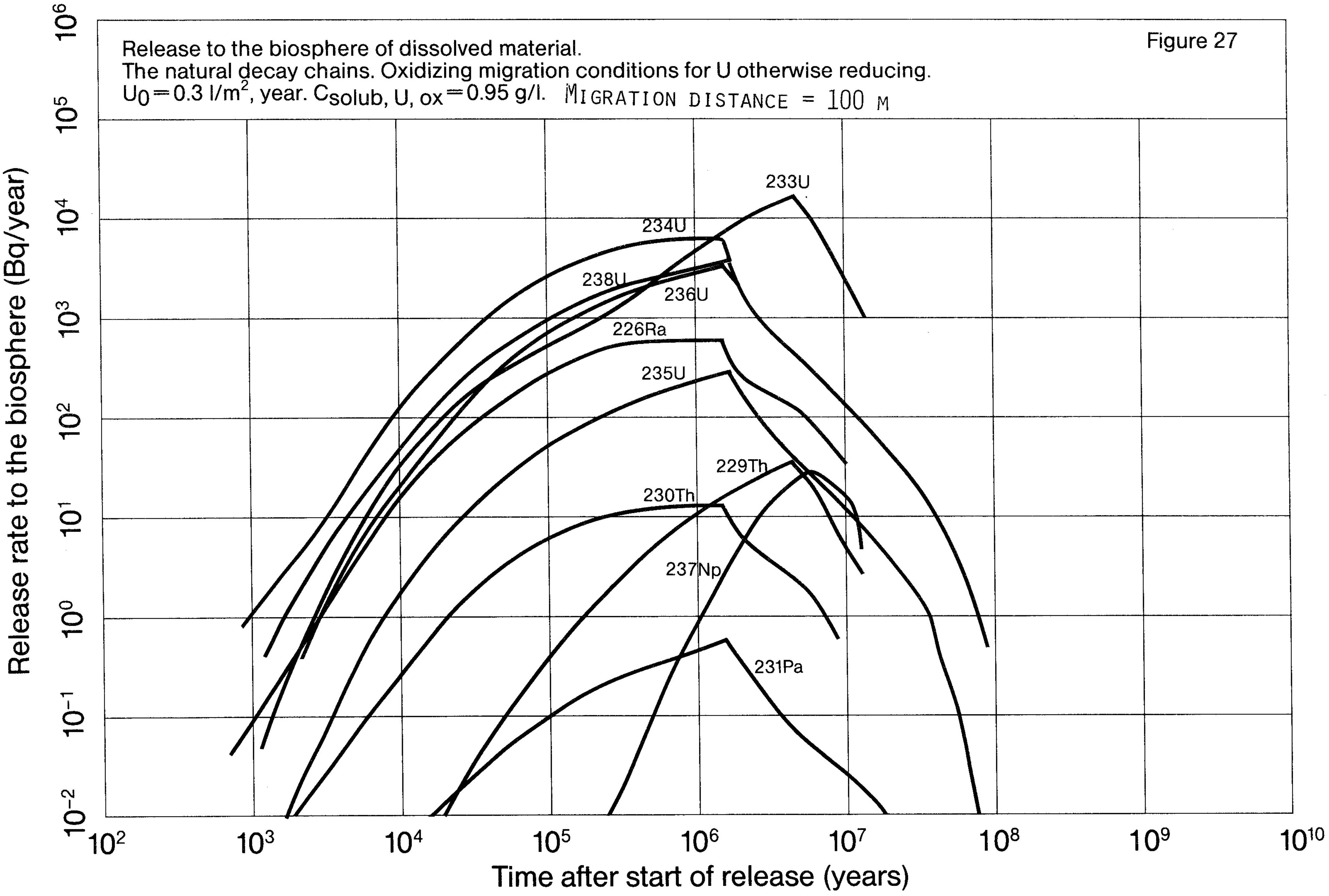


Figure 27

Release to the biosphere of dissolved material.
The natural decay chains. Oxidizing migration conditions for U otherwise reducing.
$U_0 = 0.3$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l. MIGRATION DISTANCE = 100 M

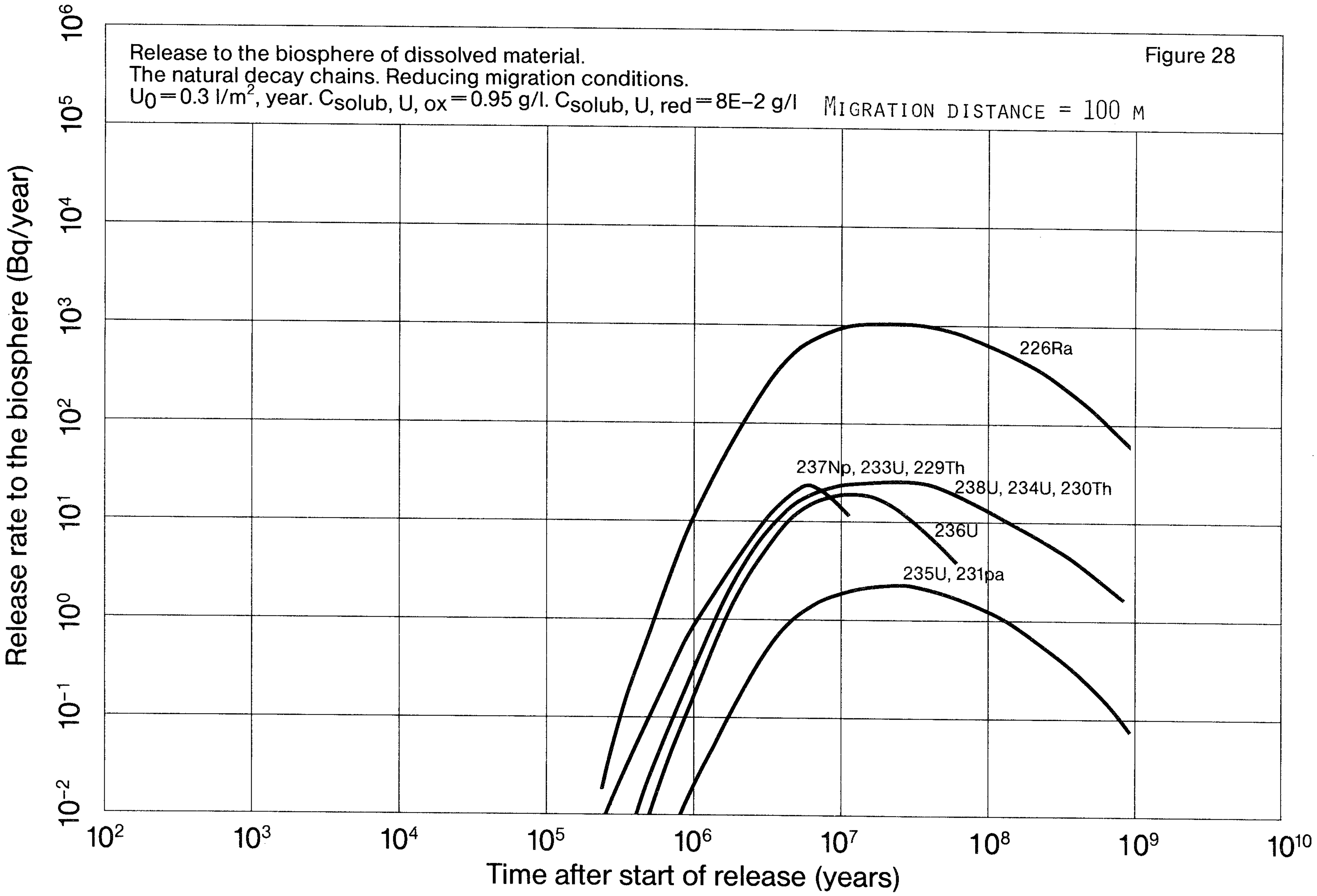


Figure 28

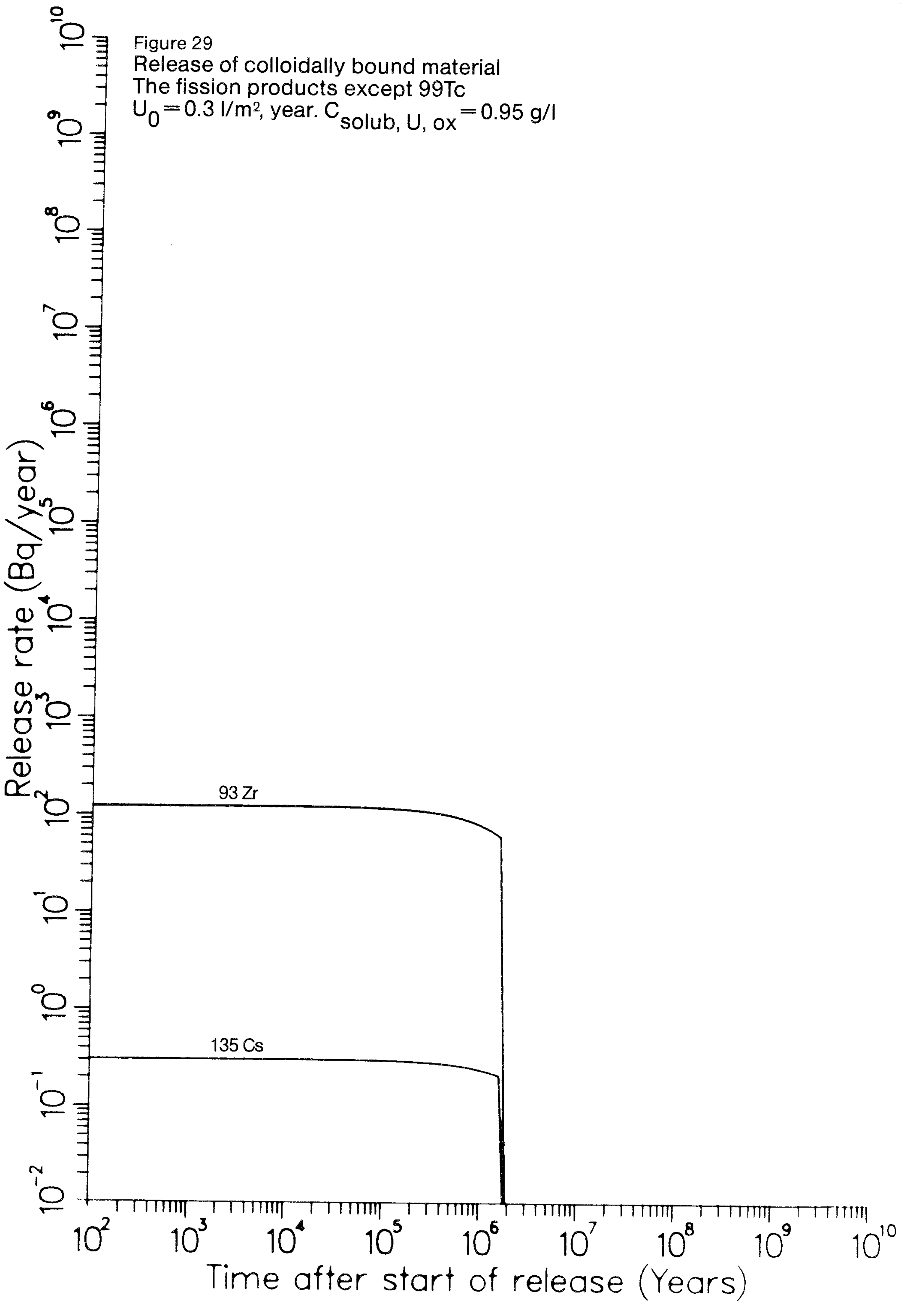
Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions.
$U_0 = 0.3$ l/m^2, year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 8E{-}2$ g/l MIGRATION DISTANCE = 100 M

Release of colloidally bound material.

$U_o=0.3$ l/m^2,year $C_{solub,U,ox}=0.95$ g/l

Figure 29	The fission products except 99Tc
Figure 30	The natural decay chains and 99 Tc from the oxidizing zone
Figure 31	The natural decay chains and 99Tc from the reducing zone.

Figure 29
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 0.3\ l/m^2$, year. $C_{solub,\ U,\ ox} = 0.95$ g/l



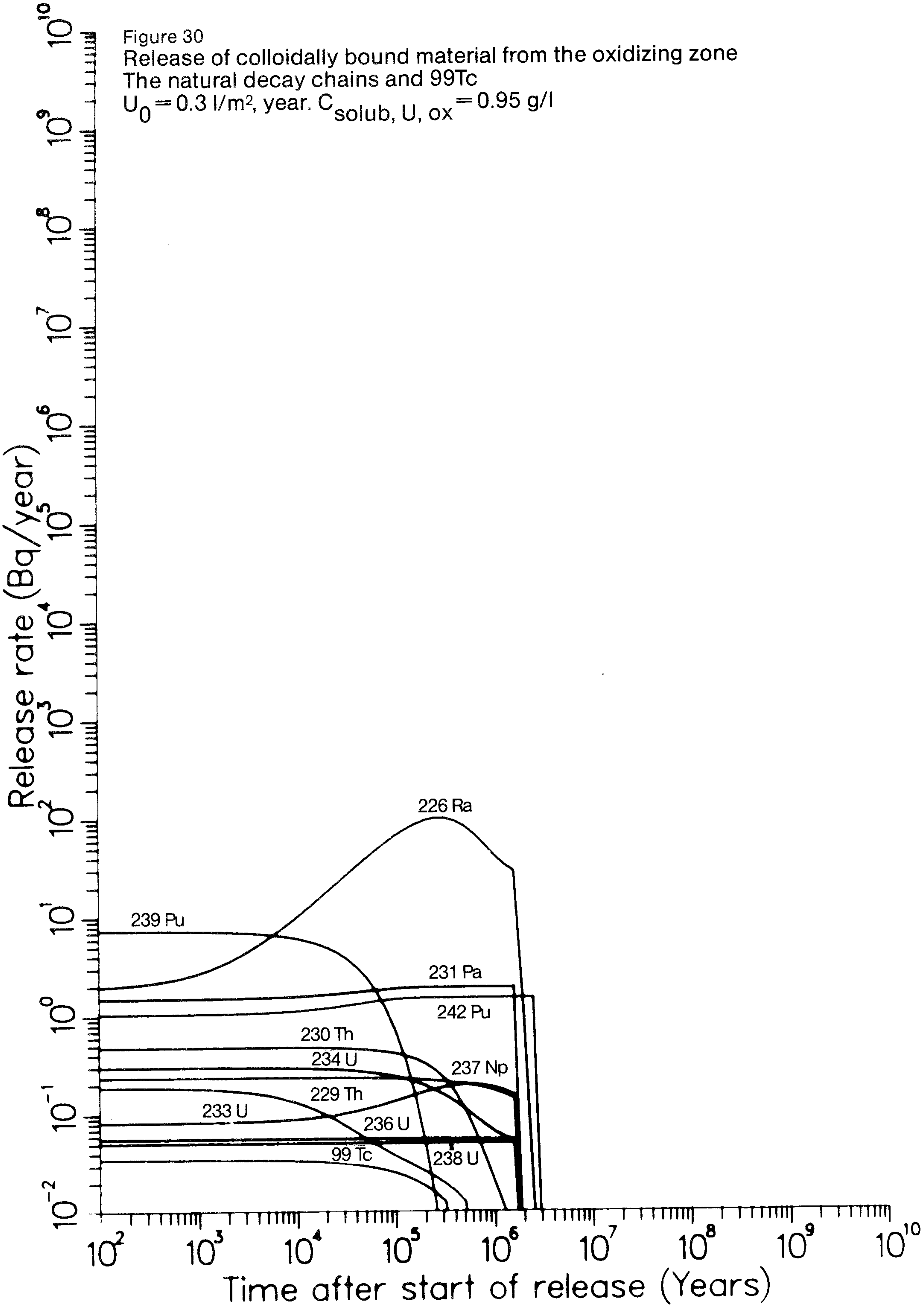


Figure 30
Release of colloidally bound material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.3$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l

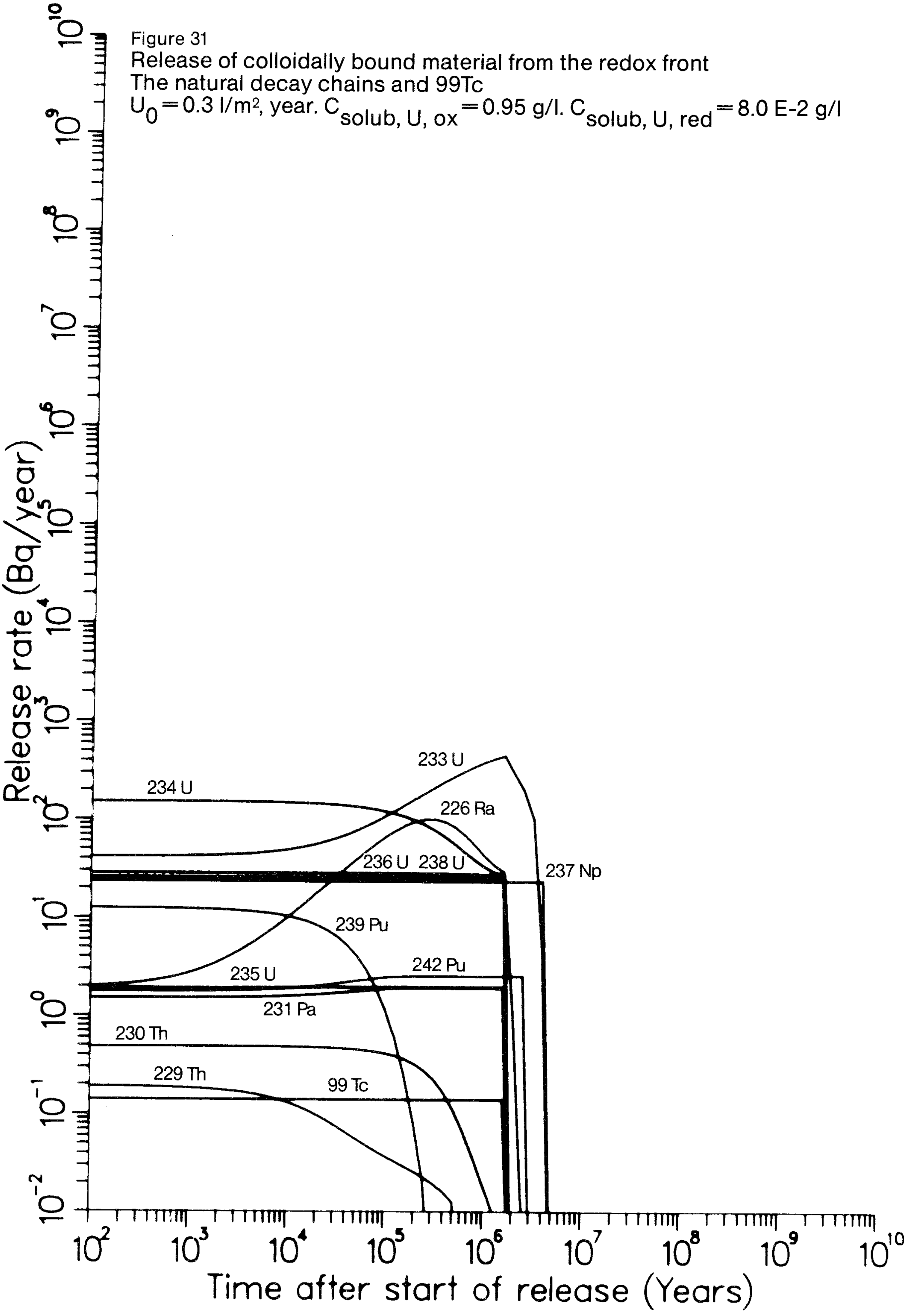


Figure 31
Release of colloidally bound material from the redox front
The natural decay chains and 99Tc
$U_0 = 0.3$ l/m², year. $C_{solub, U, ox} = 0.95$ g/l. $C_{solub, U, red} = 8.0$ E-2 g/l

Release of dissolved material to the geosphere.

U_0= 0.1 l/m^2,year $C_{solub,U,ox}$=0.36 g/l.

Figure 32	The fission products except 99Tc
Figure 33	The natural decay chains and 99Tc in oxidized form
Figure 34	The natural decay chains and 99Tc in reduced form $C_{solub,U,red}=1\cdot 10^{-5}$ g/l

Comments:

Pu and Th will initially be left behind when the fuel matrix dissolves. U, Np and Tc will precipitate at the redox front and the production of Th from solid material at the redox front will soon exceed the solubility limit so a solid Th-phase will evolve there too.

TABLE 12. Existence of solid phase (years after start of release)

Substance	In oxidized form	At the redox front
Tc	-	0-2E6
Th	0-3E9	3E4-1E9
Np	-	0-8E6
U	0-8E6	0-6E9
Pu	0-3E6	-

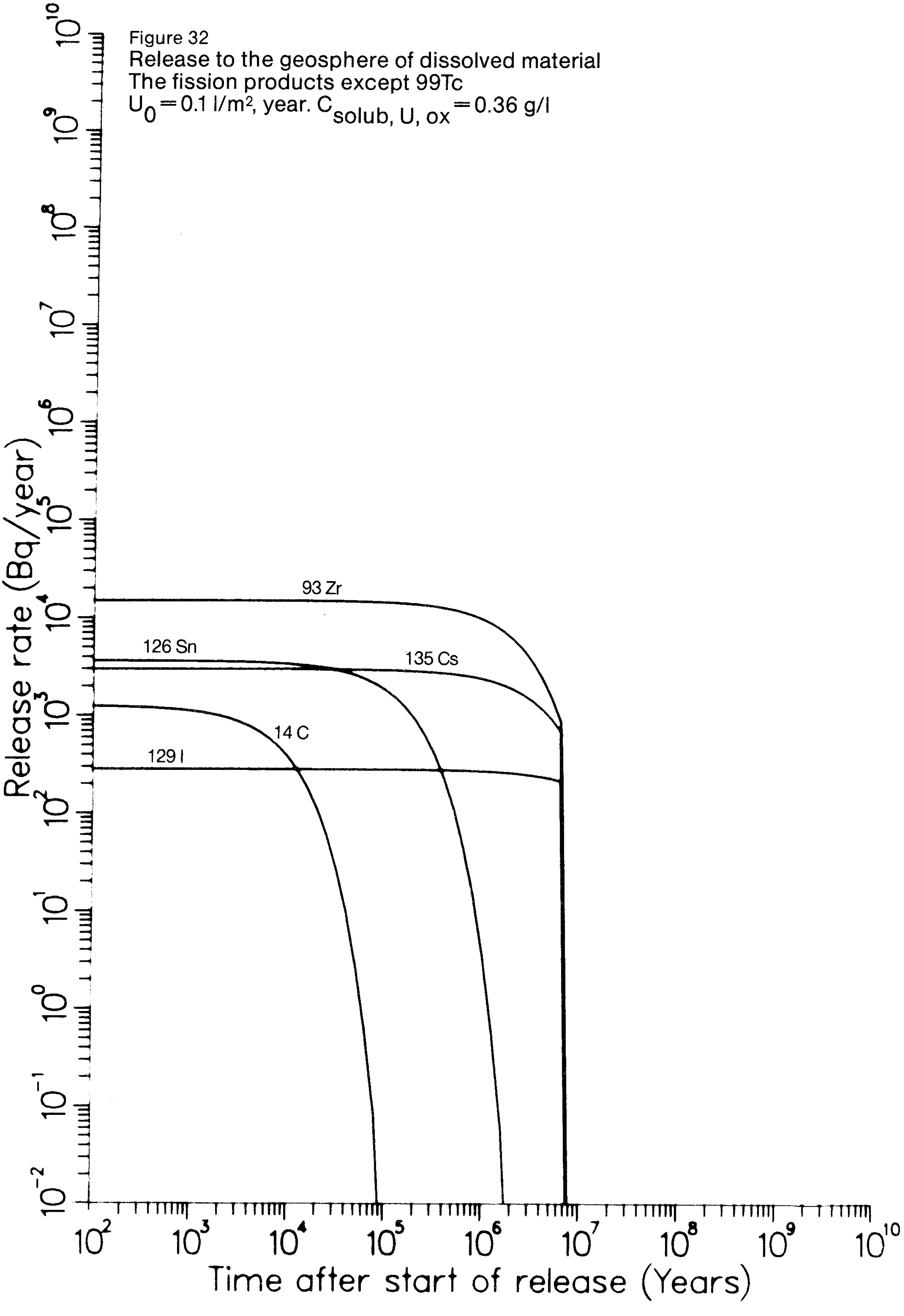


Figure 32
Release to the geosphere of dissolved material
The fission products except 99Tc
$U_0 = 0.1$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l

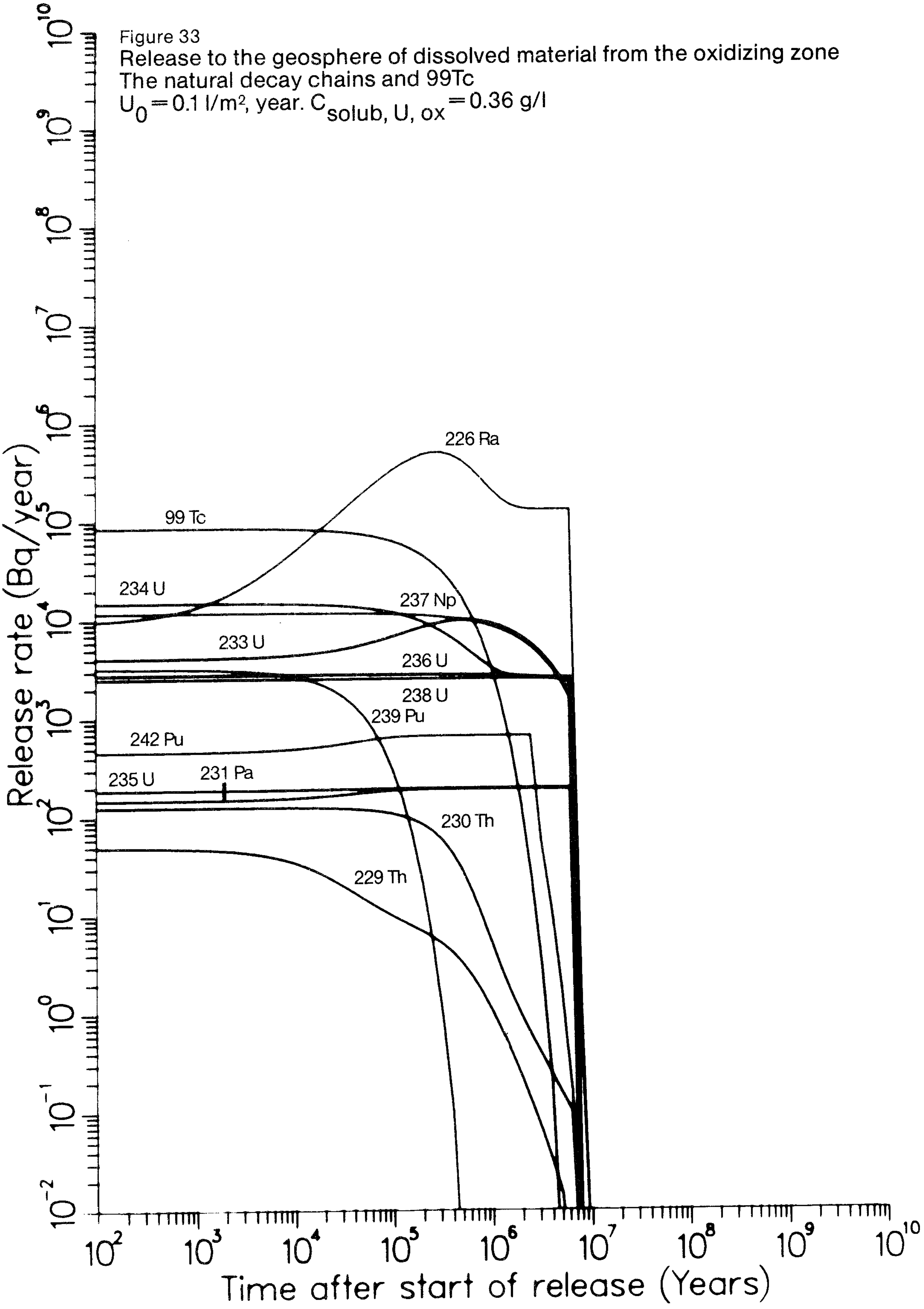


Figure 33
Release to the geosphere of dissolved material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.1\ l/m^2$, year. $C_{solub,\,U,\,ox} = 0.36\ g/l$

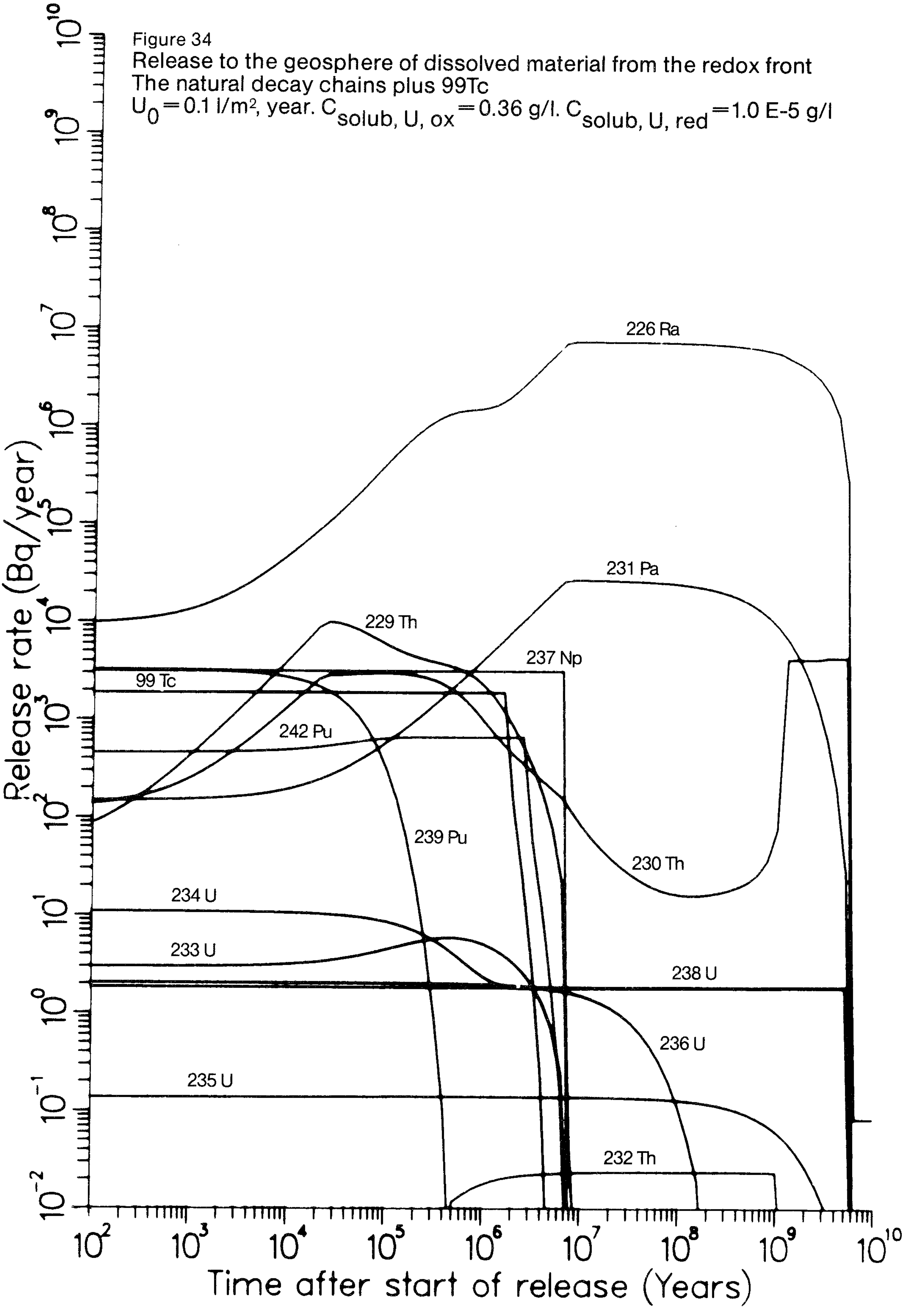


Figure 34
Release to the geosphere of dissolved material from the redox front
The natural decay chains plus 99Tc
$U_0 = 0.1$ l/m², year. $C_{solub,\,U,\,ox} = 0.36$ g/l. $C_{solub,\,U,\,red} = 1.0$ E-5 g/l

Release of dissolved material to the biosphere.
U_o=0.1 l/m^2,year $C_{solub,U,ox}$=0.36 g/l migration distance =100 m

Figure 35 The fission products

Figure 36 The natural decay chains. Oxidizing migration conditions

Figure 37 The natural decay chains. Reducing migration conditions. $C_{solub,U,red}=1 \cdot 10^{-5}$ g/l

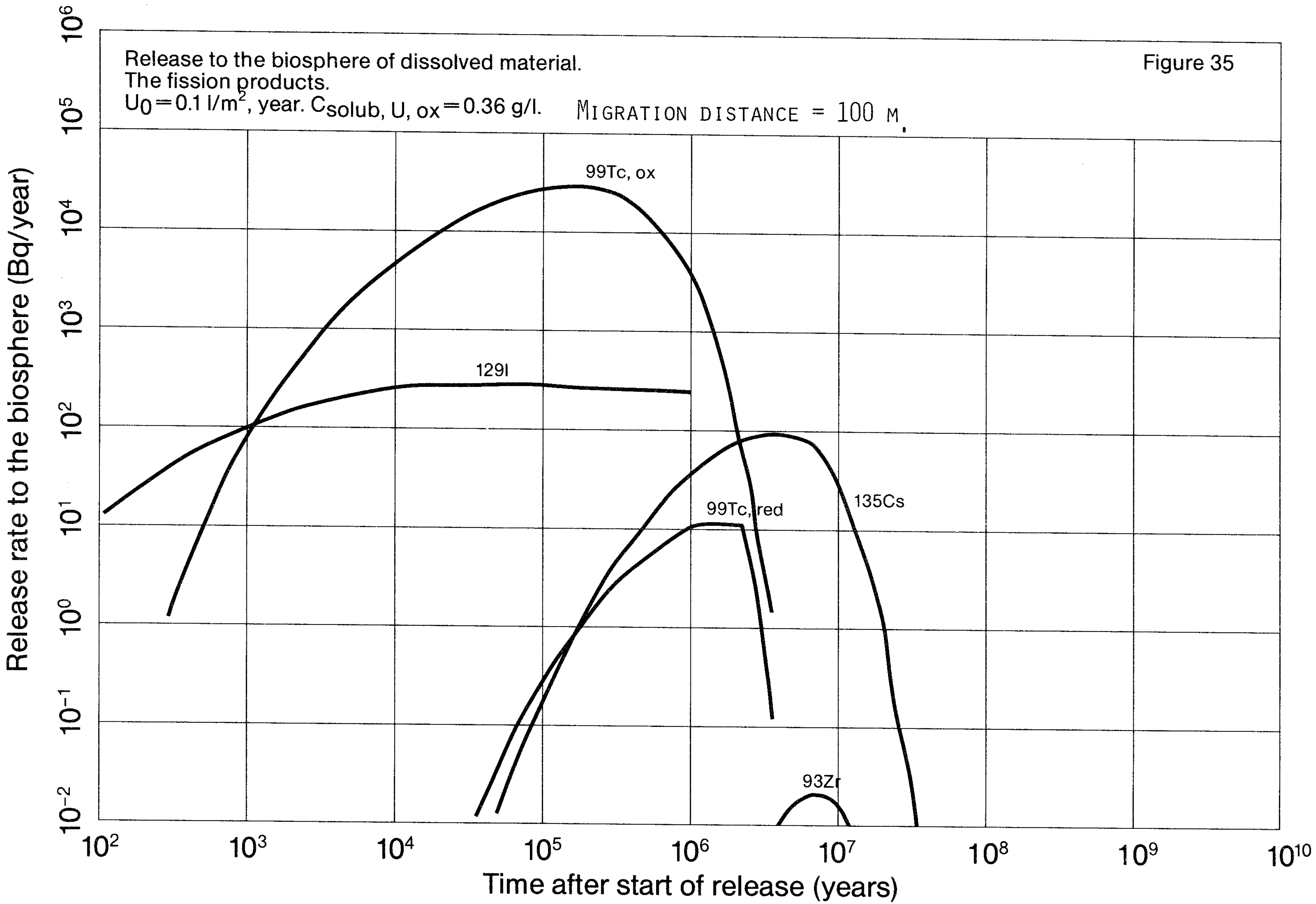


Figure 35

Release to the biosphere of dissolved material.
The fission products.
$U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l. MIGRATION DISTANCE = 100 M.

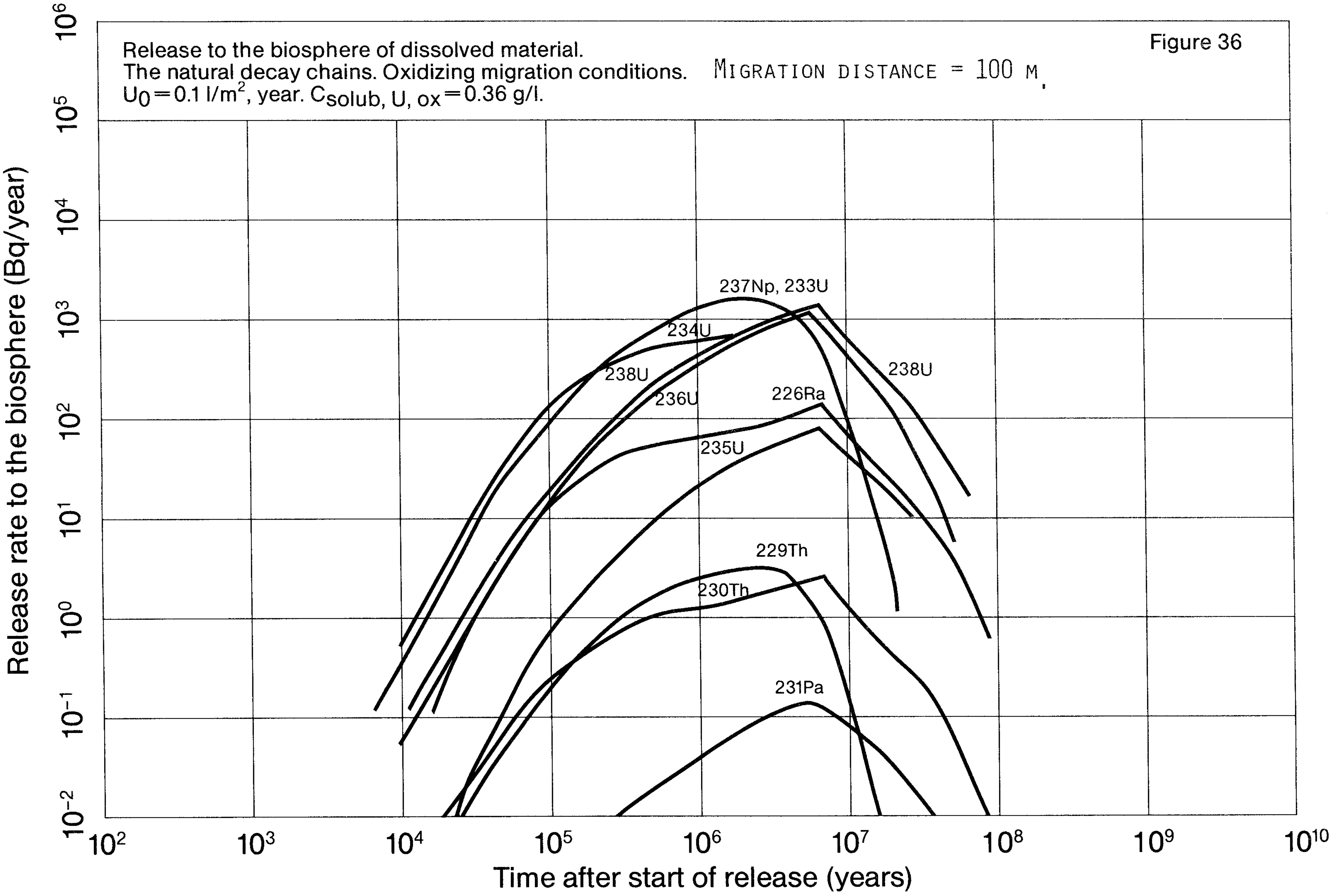


Figure 36
Release to the biosphere of dissolved material.
The natural decay chains. Oxidizing migration conditions.
MIGRATION DISTANCE = 100 M.
$U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l.
Release rate to the biosphere (Bq/year)
10^{-2}
10^{-1}
10^0
10^1
10^2
10^3
10^4
10^5
10^6
Time after start of release (years)
10^2
10^3
10^4
10^5
10^6
10^7
10^8
10^9
10^{10}
237Np, 233U
234U
238U
236U
238U
226Ra
235U
229Th
230Th
231Pa

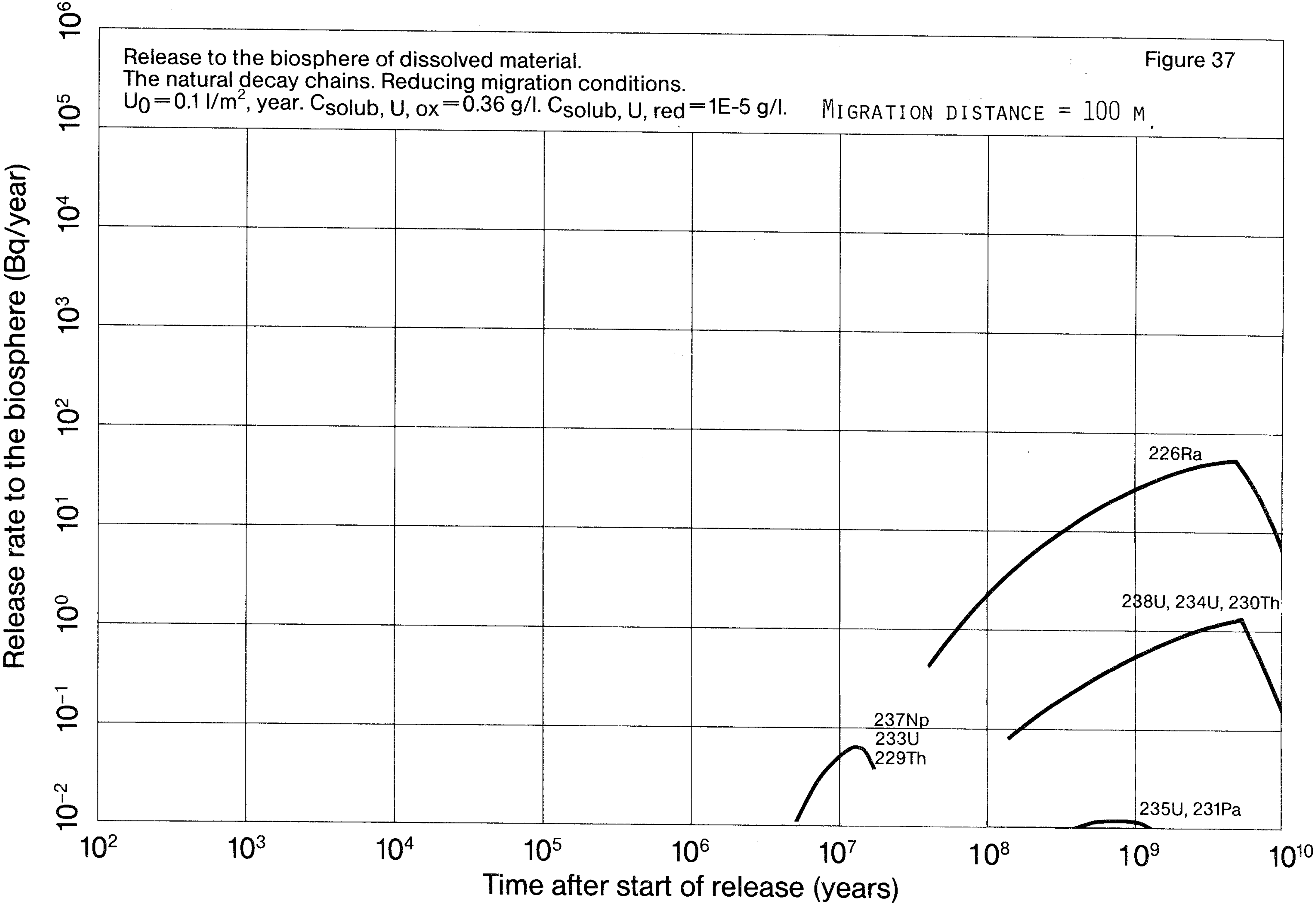


Figure 37

Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions.
$U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l. $C_{solub, U, red} = 1E\text{-}5$ g/l. MIGRATION DISTANCE = 100 M.

Release of colloidally bound material.

U_o=0.1 l/m^2,year $C_{solub,U,ox}$=0.36 g/l

$C_{solub,U,red}$=$1\cdot10^{-5}$ g/l.

Figure 38	The fission products except 99Tc
Figure 39	The natural decay chains and 99Tc in oxidized form
Figure 40	The natural decay chains and 99Tc in reduced form.

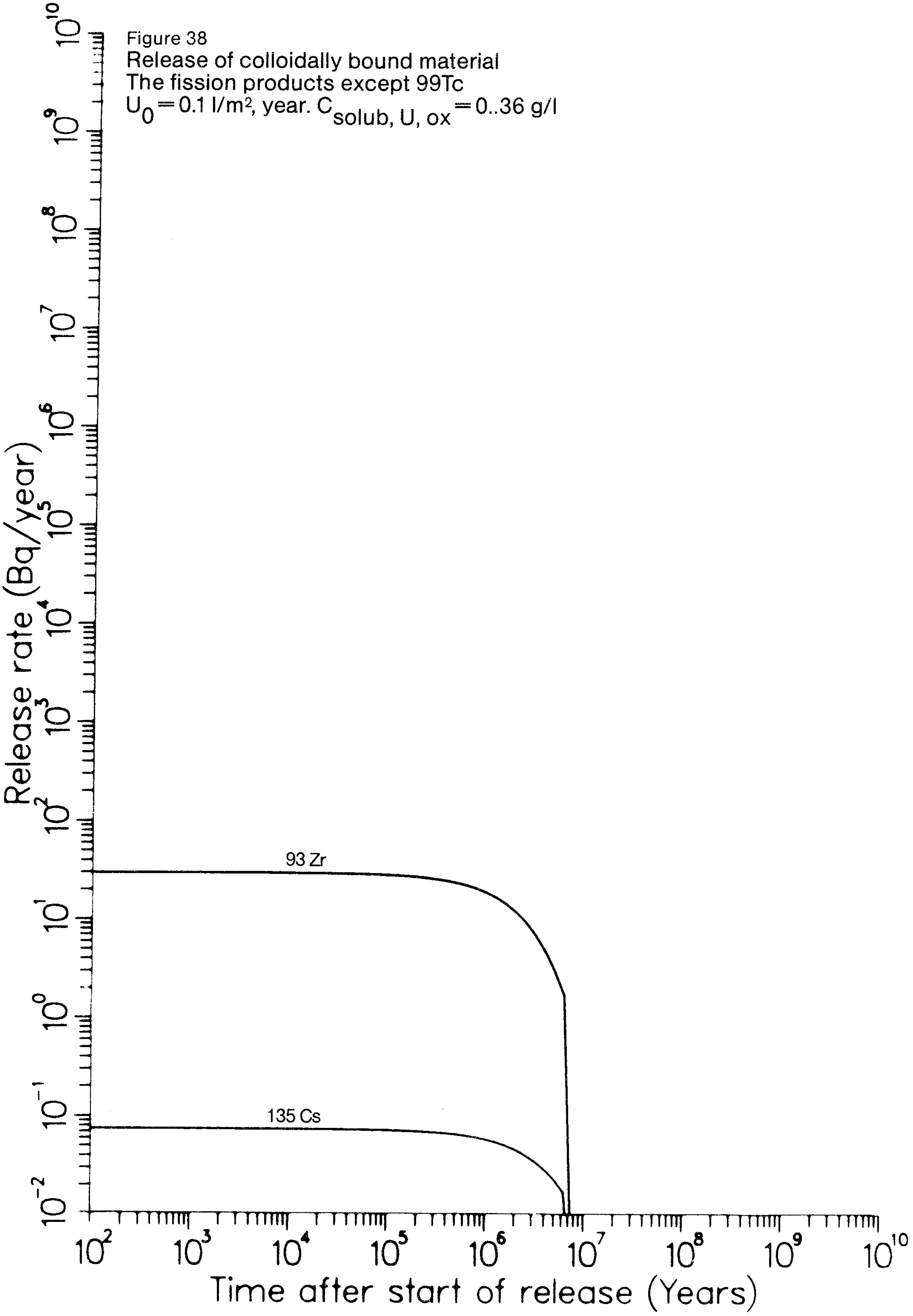


Figure 38
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0..36$ g/l

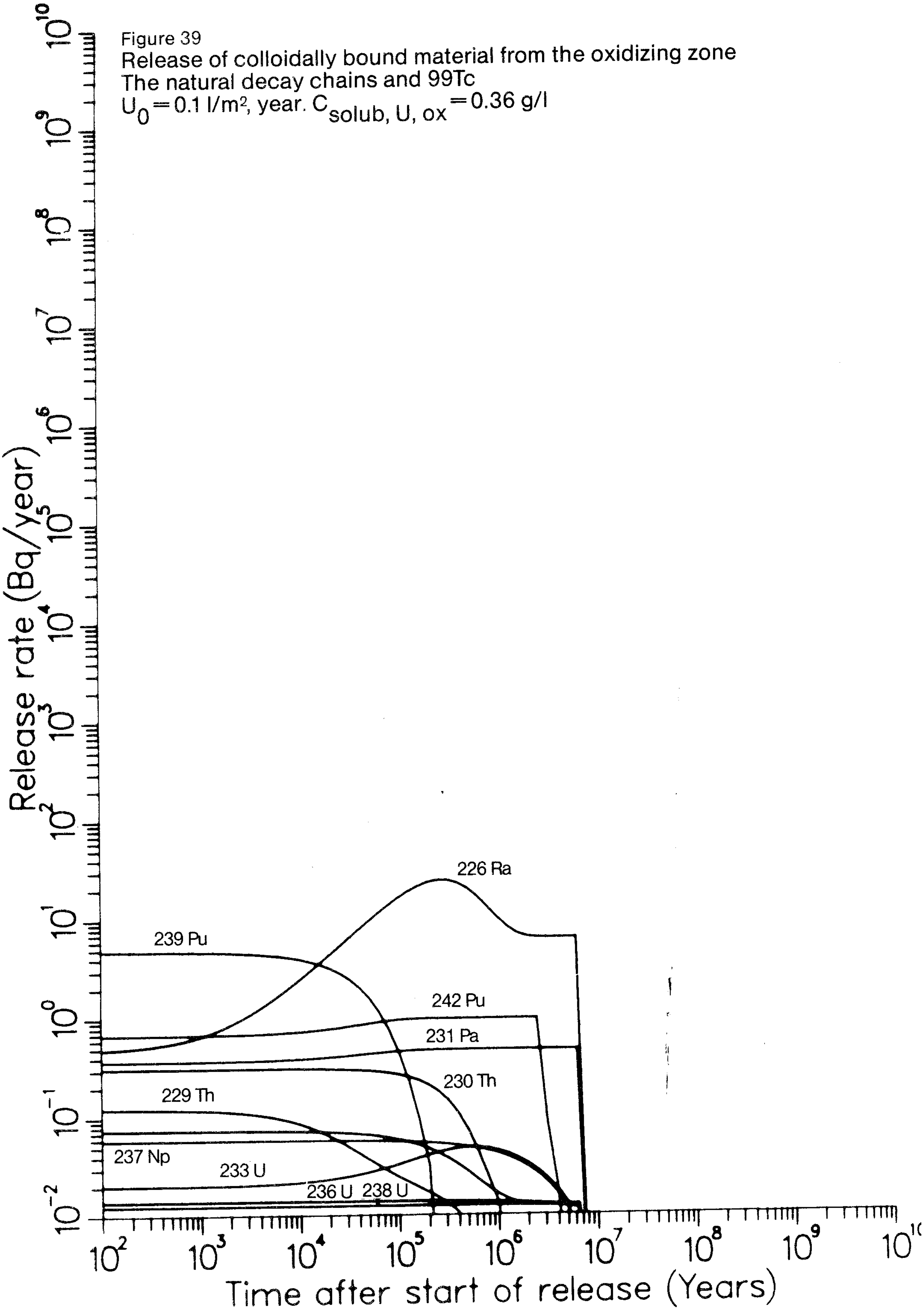


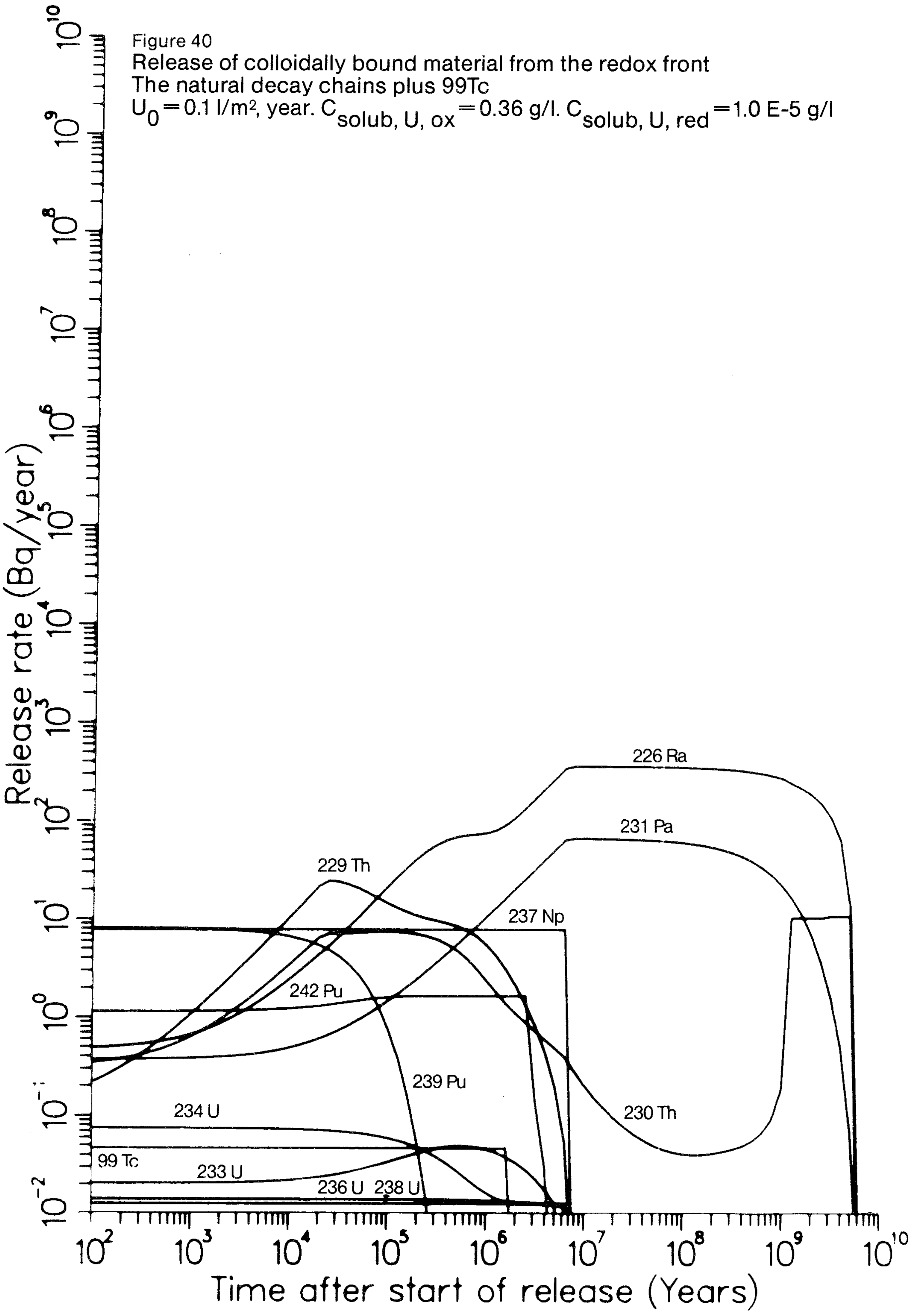
Figure 39

Release of colloidally bound material from the oxidizing zone

The natural decay chains and 99Tc

$U_0 = 0.1\ l/m^2$, year. $C_{solub, U, ox} = 0.36$ g/l

Figure 40
Release of colloidally bound material from the redox front
The natural decay chains plus 99Tc
$U_0 = 0.1\ l/m^2$, year. $C_{solub, U, ox} = 0.36$ g/l. $C_{solub, U, red} = 1.0$ E-5 g/l



Release of dissolved material to the geosphere. U_0= 0.1 l/m^2,year $C_{solub,U,ox}$=0.36 g/l initially defective canister.

Figure 41 The fission products except 99Tc

Figure 42 The natural decay chains and 99Tc from the oxidizing zone

Figure 43 The natural decay chains and 99Tc from the redox front. $C_{solub,U,red}=1\cdot10^{-5}$ g/l

TABLE 13. Existence of solid phase (years after start of leaching)

Substance	The oxidizing zone	The redox front
Tc	-	0-2E6
Th	0-2E9	2E4-2E9
Np	-	0-8E6
U	0-5E6	0-6E9
Pu	0-3E6	-

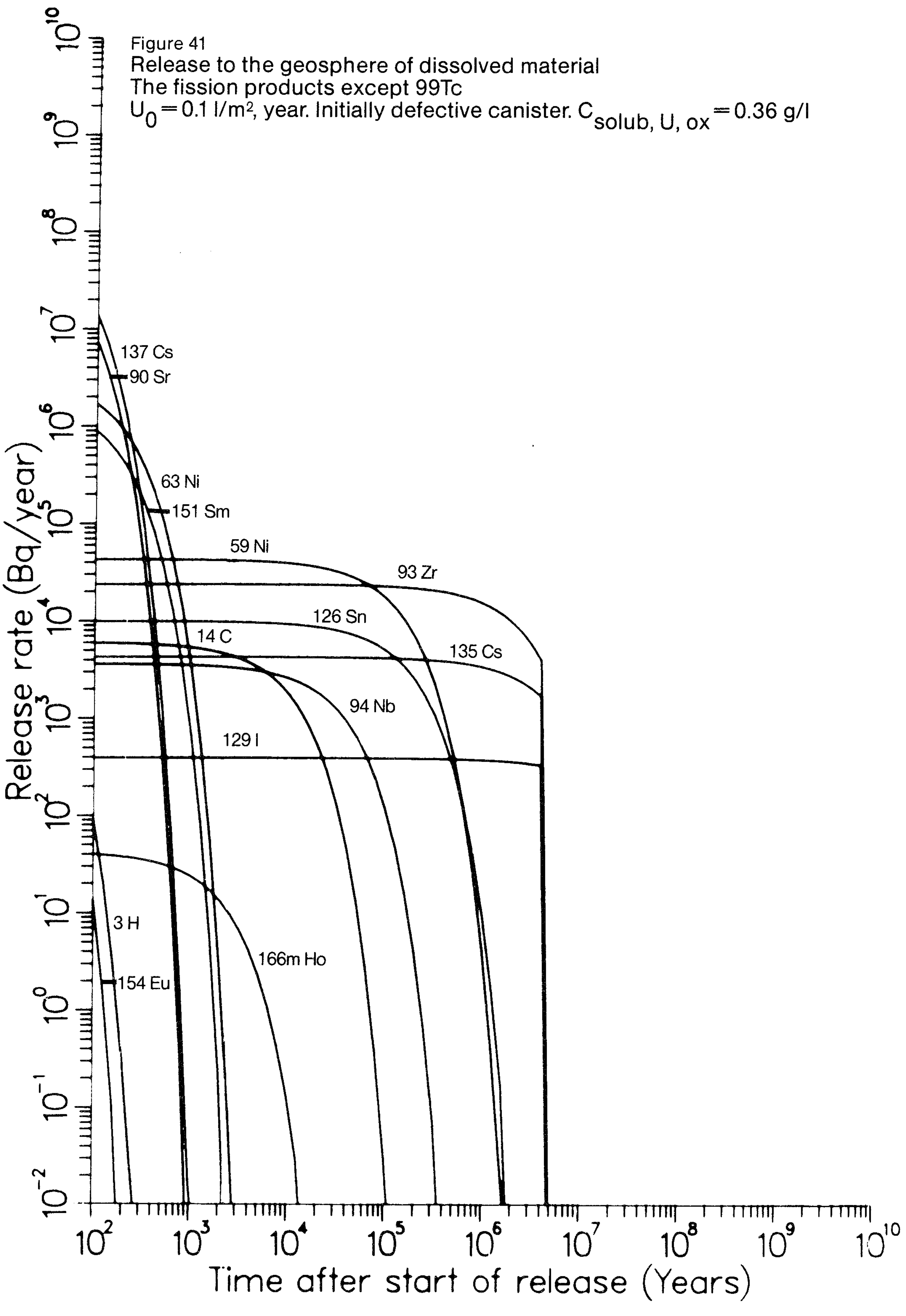


Figure 41
Release to the geosphere of dissolved material
The fission products except 99Tc
$U_0 = 0.1\ \mathrm{l/m^2}$, year. Initially defective canister. $C_{solub,\ U,\ ox} = 0.36$ g/l

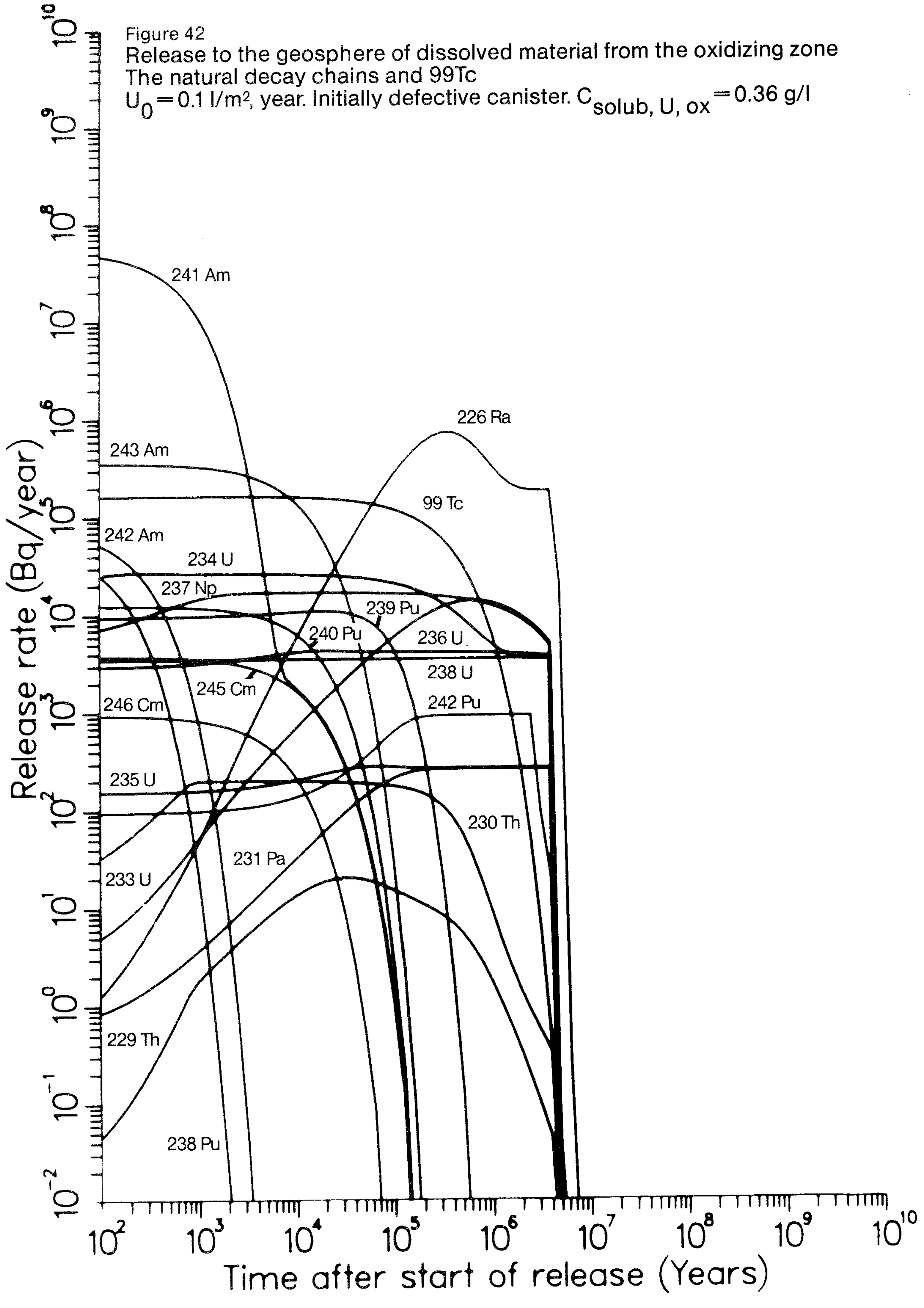


Figure 42
Release to the geosphere of dissolved material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.1$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.36$ g/l

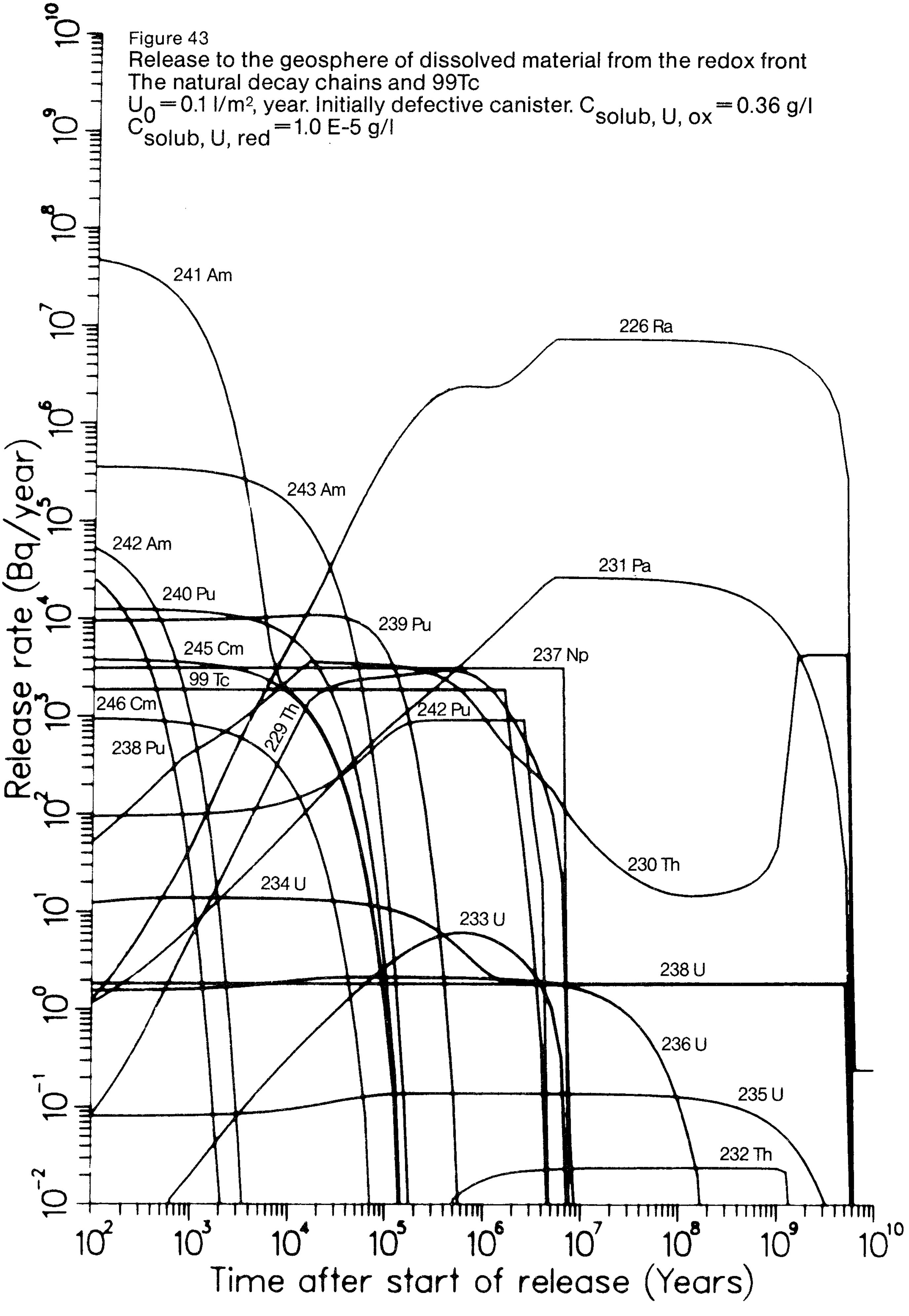


Figure 43
Release to the geosphere of dissolved material from the redox front
The natural decay chains and 99Tc
$U_0 = 0.1$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.36$ g/l
$C^0_{solub, U, red} = 1.0$ E-5 g/l

Release of dissolved material to the biosphere.
U_o=0.1 $1/m^2$,year $C_{solub,U,ox}$=0.36 g/l
Initially defective canister, migration distance=100 m.

Figure 44	The fission products
Figure 45	The natural decay chains. Oxidizing migration conditions
Figure 46	The natural decay chains, Reducing migration conditions. $C_{solub,U,red}=1\cdot10^{-5}$ g/l

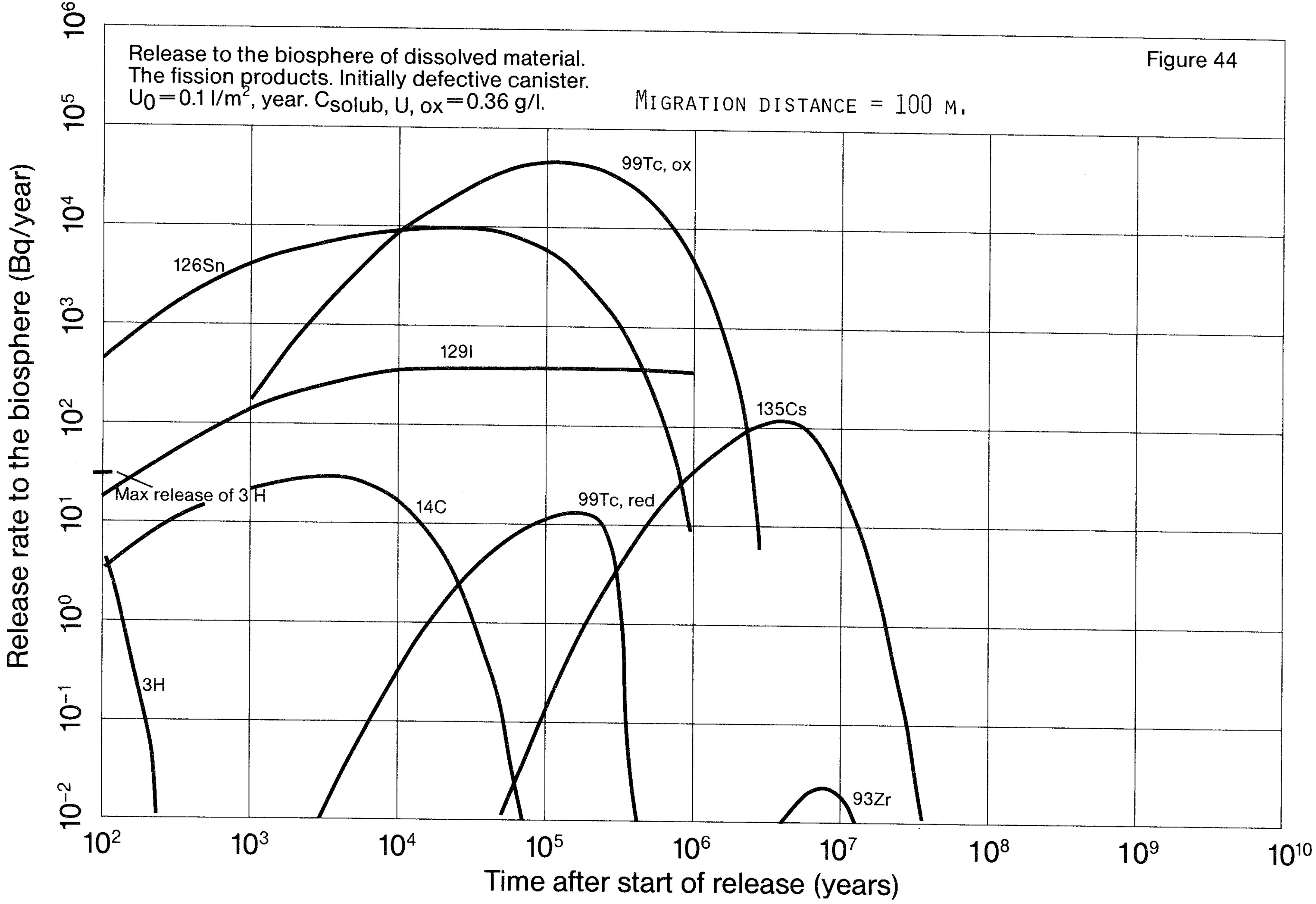


Figure 44

Release to the biosphere of dissolved material. The fission products. Initially defective canister. $U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l.

MIGRATION DISTANCE = 100 M.

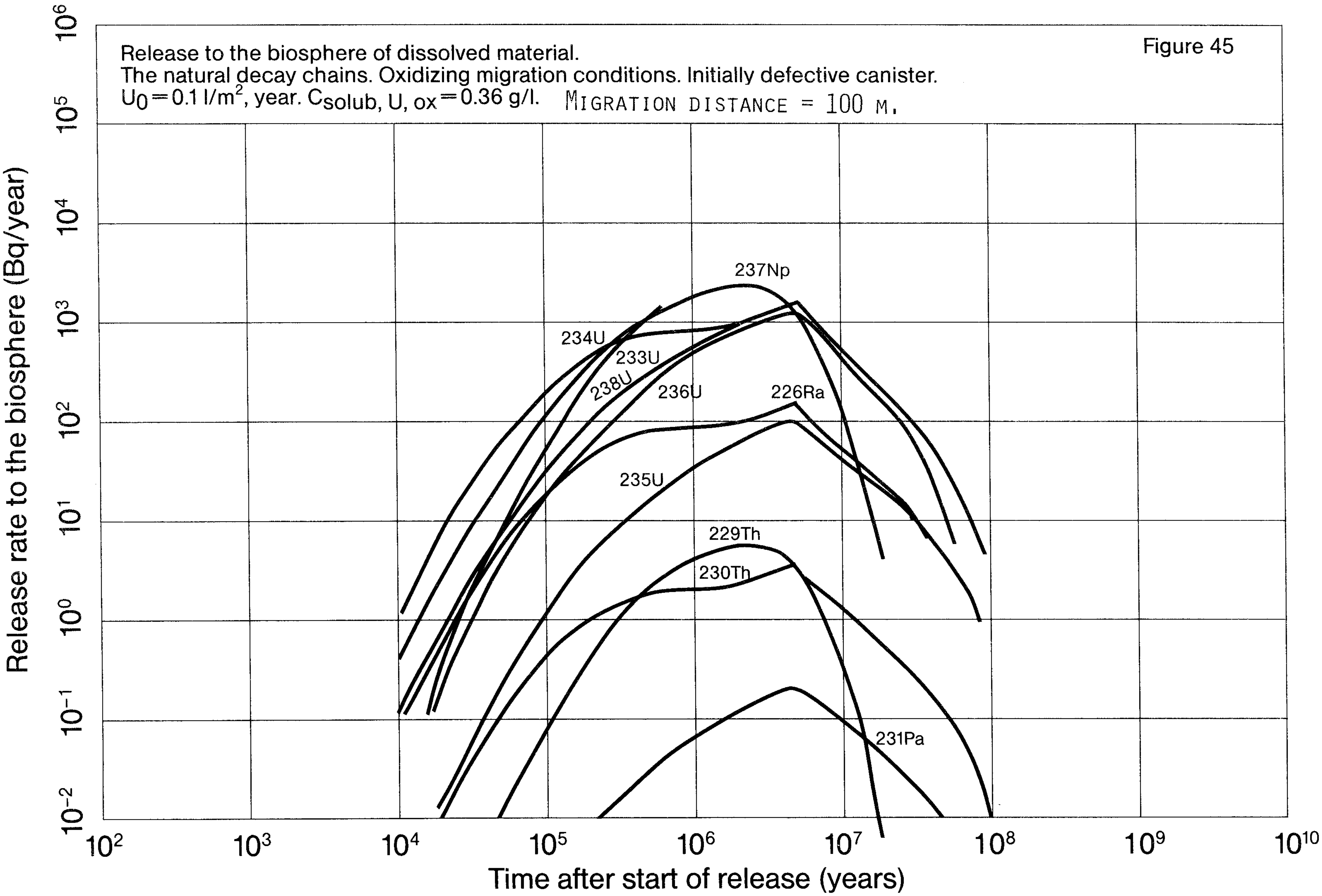
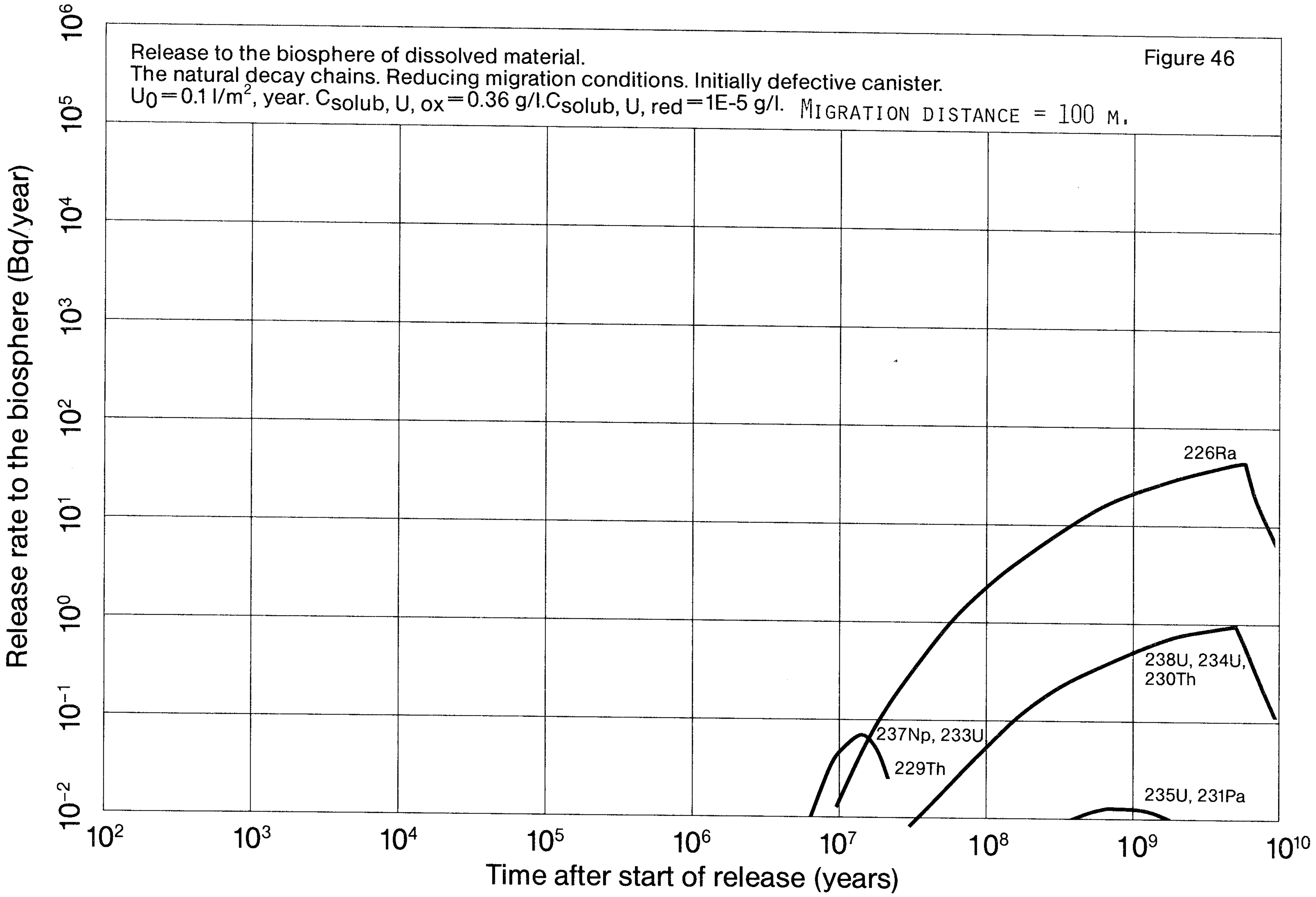


Figure 45

Release to the biosphere of dissolved material.
The natural decay chains. Oxidizing migration conditions. Initially defective canister.
$U_0 = 0.1$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l. MIGRATION DISTANCE = 100 M.



Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions. Initially defective canister.
$U_0 = 0.1$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l.$C_{solub, U, red} = 1E\text{-}5$ g/l. MIGRATION DISTANCE = 100 M.

Figure 46

Release of colloidally bound material.
U_o=0.1 l/m^2,year $C_{solub,U,ox}$= 0.36 g/l
Initially defective canister.

Figure 47	The fission products except 99Tc
Figure 48	The natural decay chains and 99Tc from the oxidizing zone
Figure 49	The natural decay chains and 99Tc from the redox front

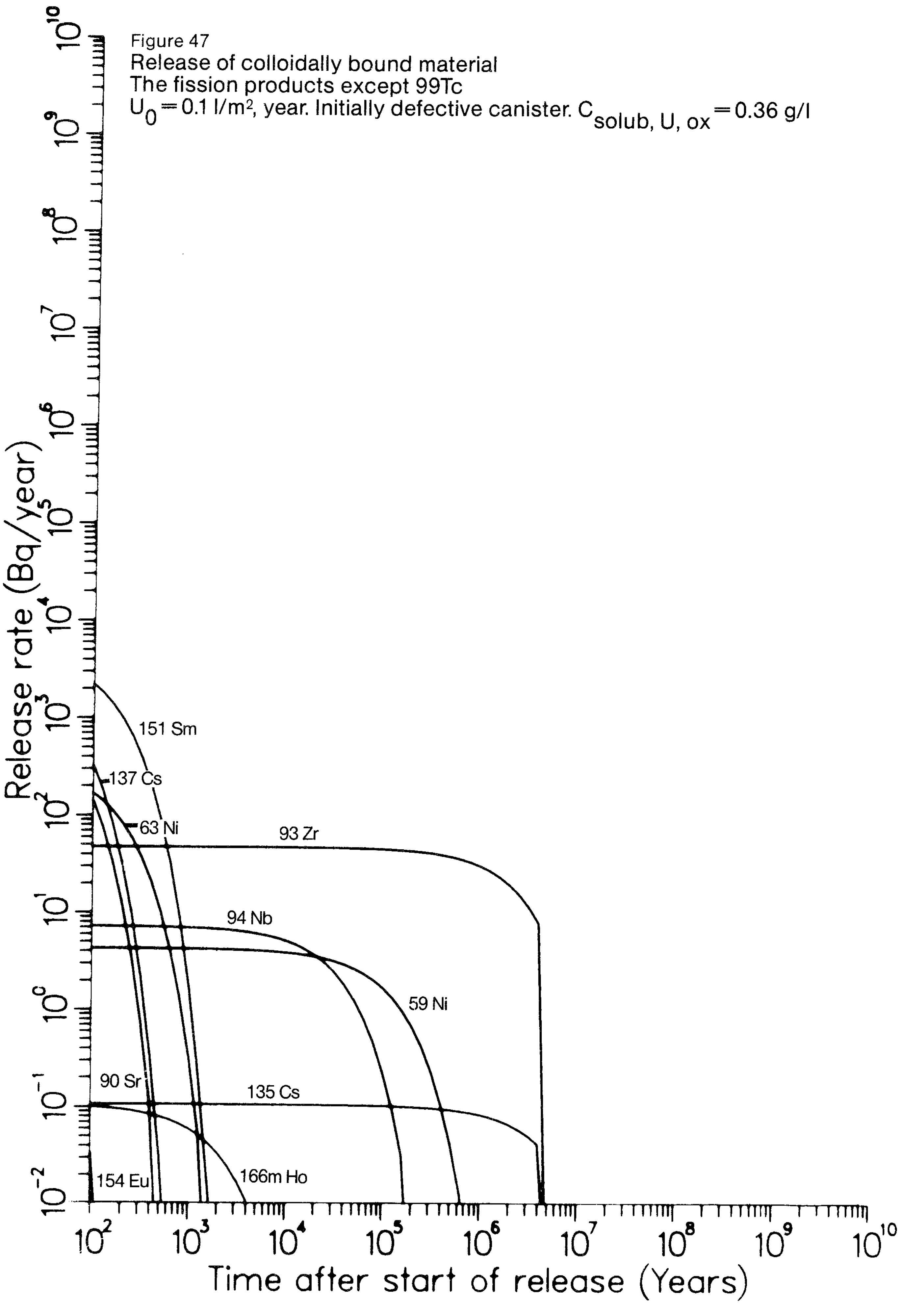


Figure 47
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 0.1$ l/m^2, year. Initially defective canister. $C_{solub, U, ox} = 0.36$ g/l

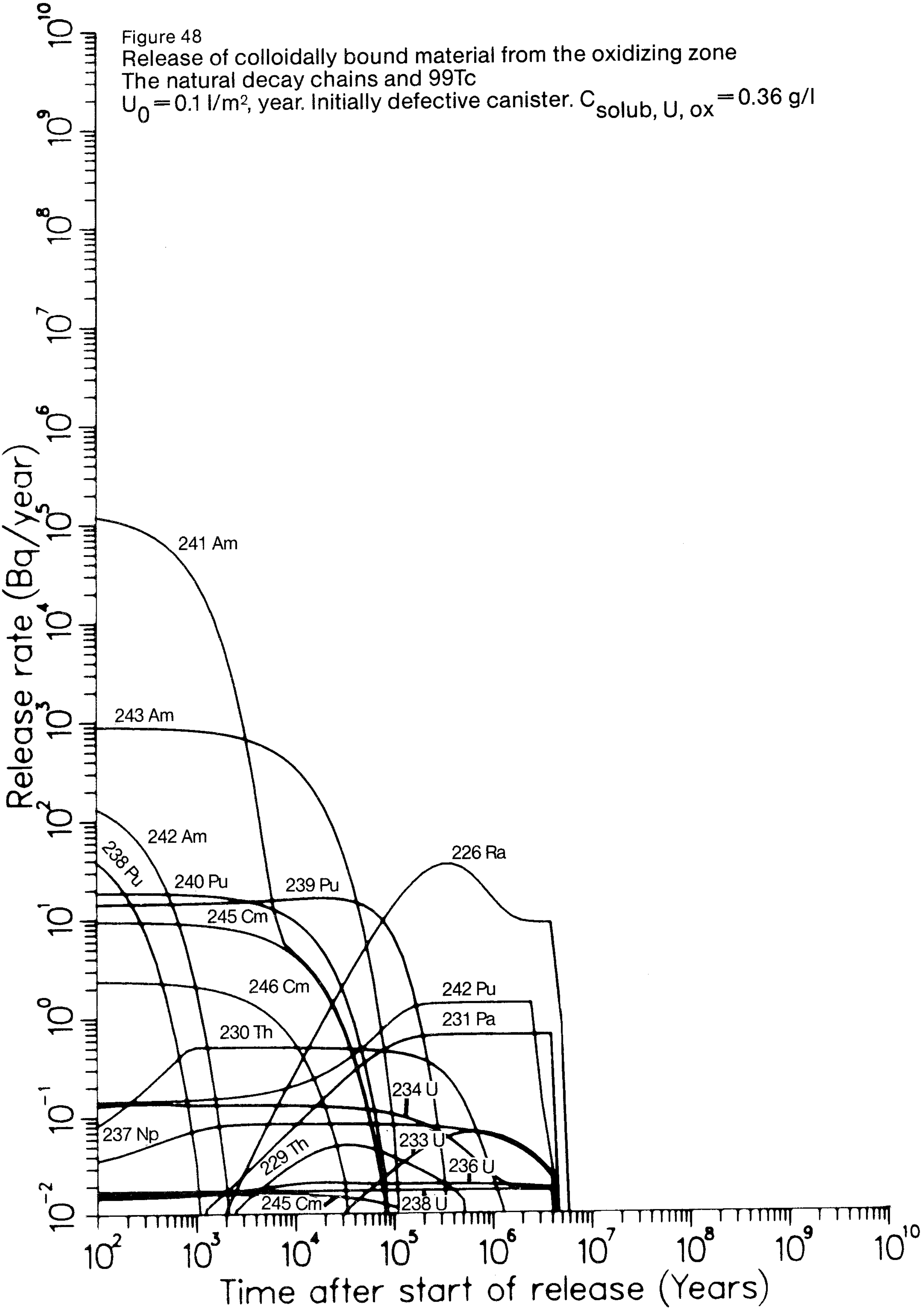


Figure 48
Release of colloidally bound material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.1$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.36$ g/l

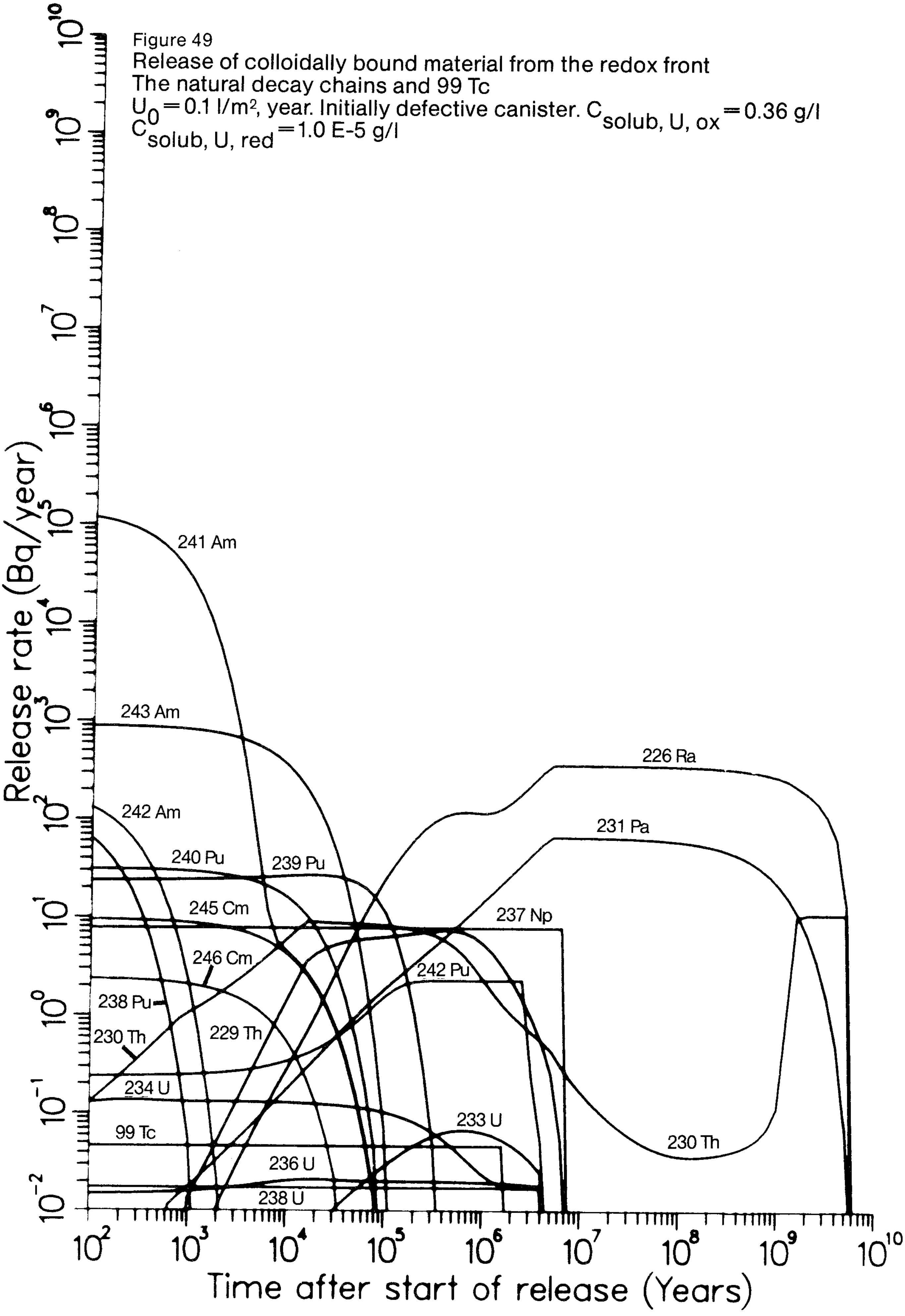


Figure 49
Release of colloidally bound material from the redox front
The natural decay chains and 99 Tc
$U_0 = 0.1$ l/m², year. Initially defective canister. $C_{solub, U, ox} = 0.36$ g/l
$C_{solub, U, red} = 1.0$ E-5 g/l

Release of dissolved material to the geosphere.
U_o=0.03 l/m^2,year $C_{solub,U,ox}$= 0.36 g/l

Figure 50 The fission products except 99Tc
Figure 51 The natural decay chains and 99Tc from the oxidizing zone
Figure 52 The natural decay chains and 99Tc from the redox front, $C_{solub,U,red}=1 \cdot 10^{-5}$ g/l.

TABLE 14. Existence of solid phase (years after start of leaching)

Substance	The oxidizing zone	The redox front
Tc	-	0-2E6
Th	0-8E9	1E4-1E10
Np	-	0-1E7
U	0-1E7	0-1E10
Pu	0-3E6	-

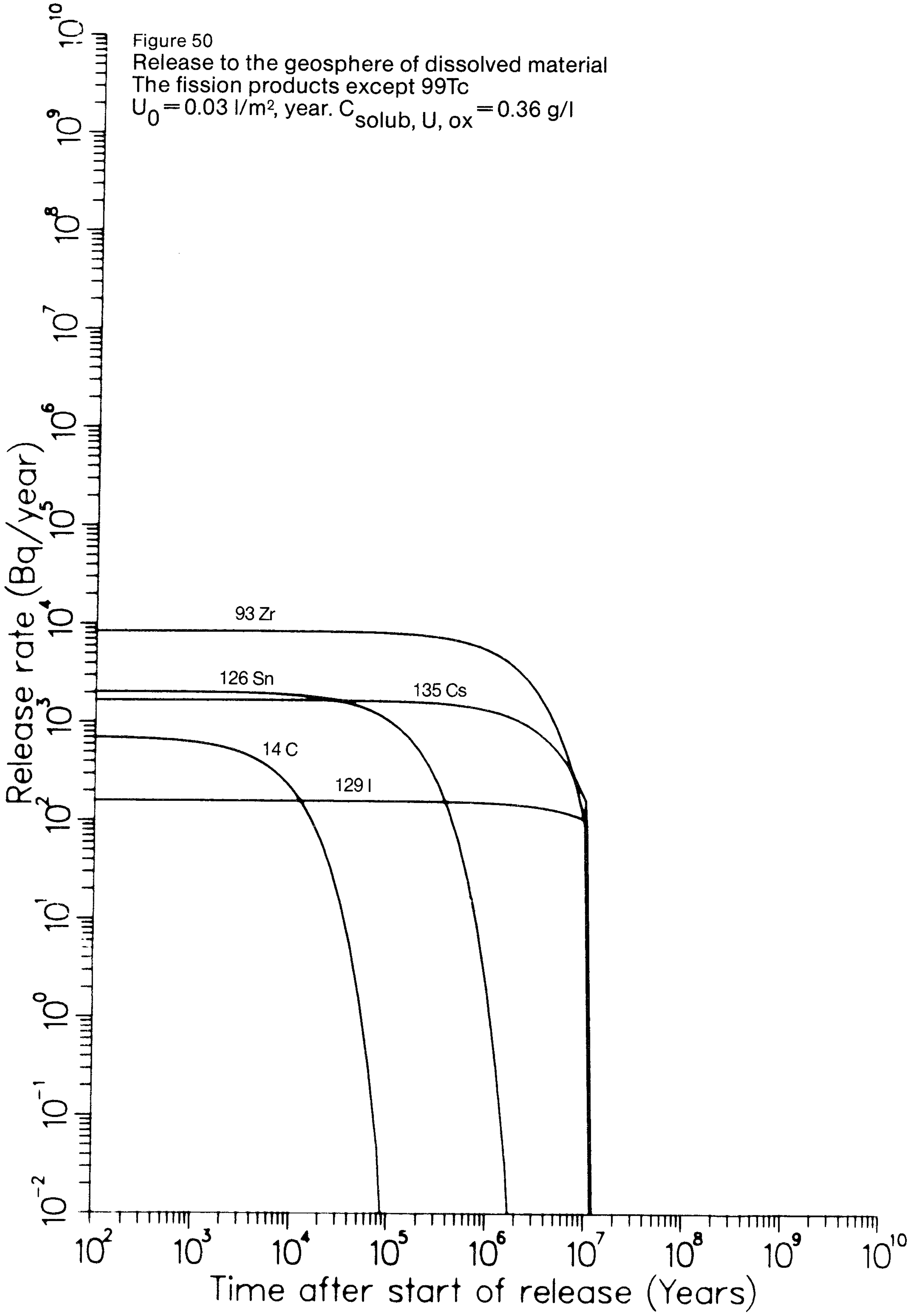


Figure 50
Release to the geosphere of dissolved material
The fission products except 99Tc
$U_0 = 0.03$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l

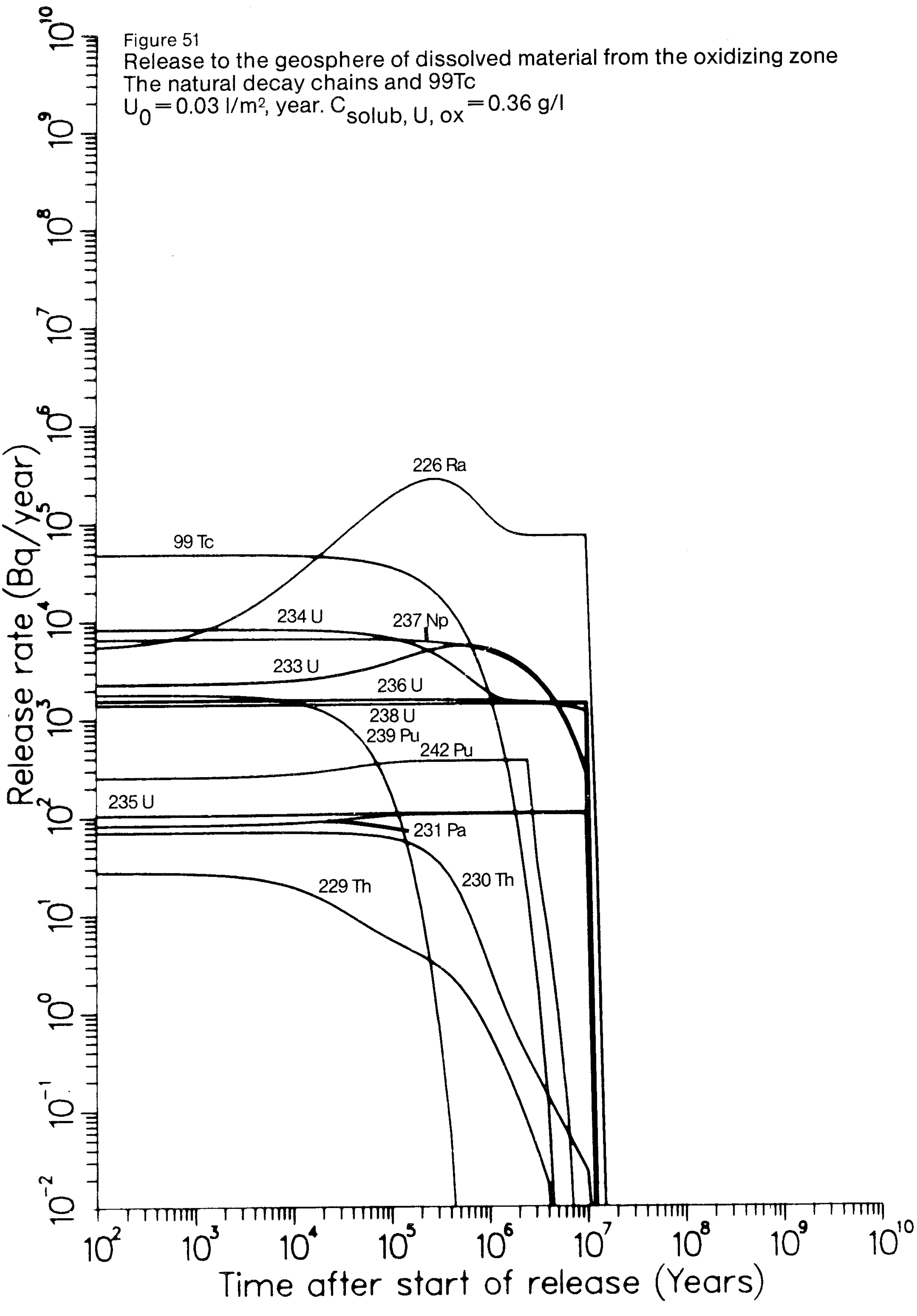
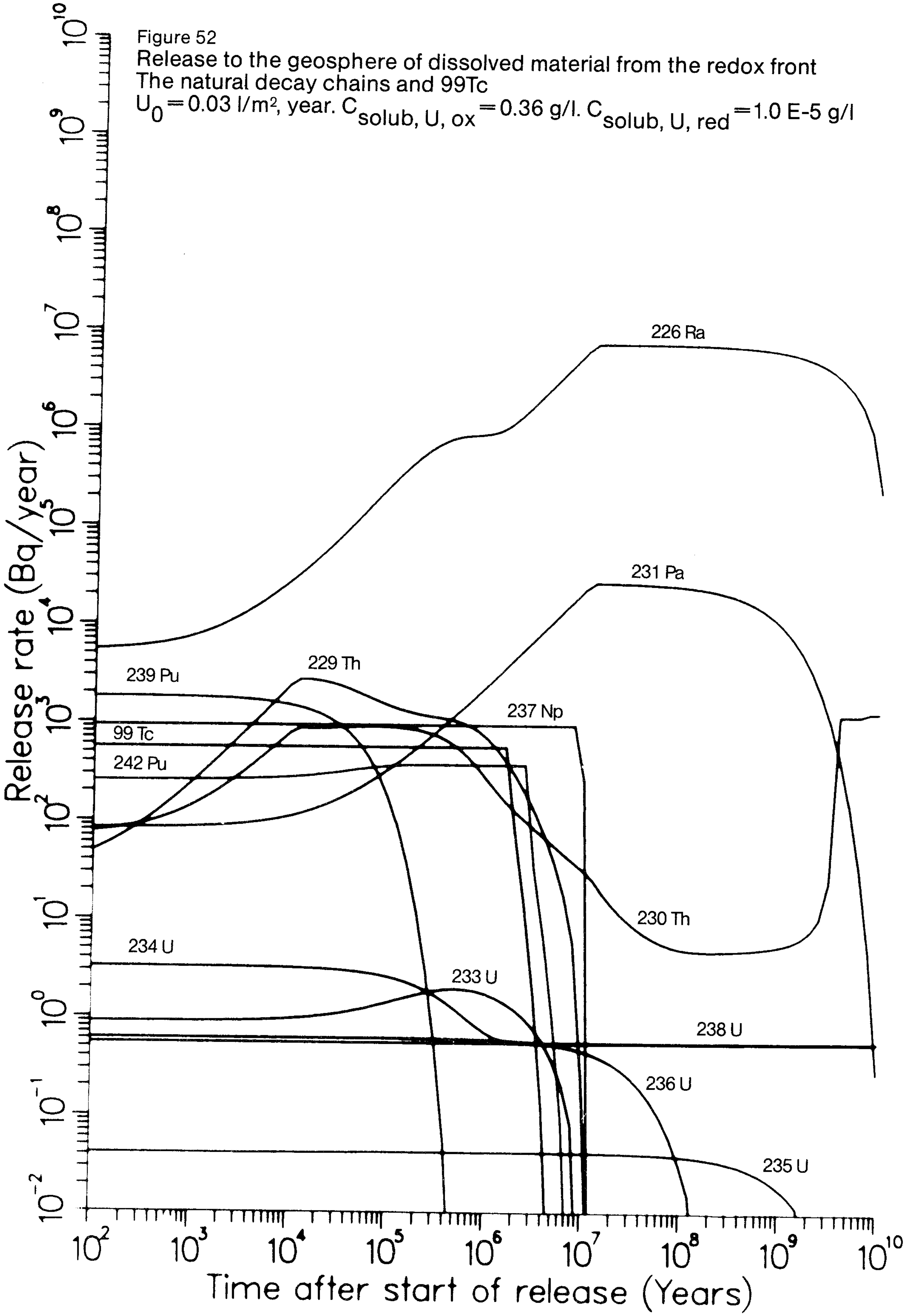


Figure 51
Release to the geosphere of dissolved material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.03$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l

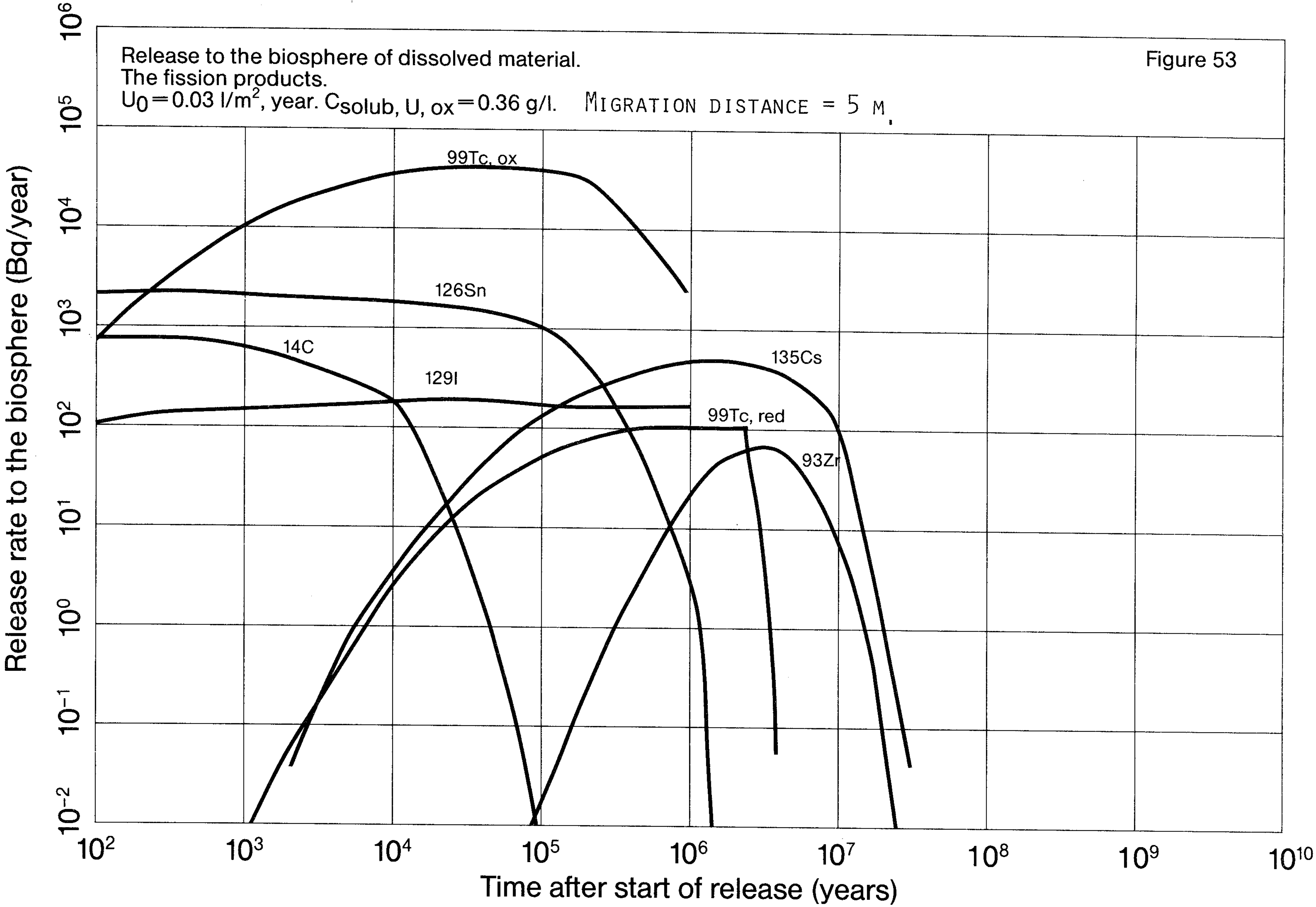
Figure 52

Release to the geosphere of dissolved material from the redox front
The natural decay chains and 99Tc
$U_0 = 0.03$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l. $C_{solub, U, red} = 1.0$ E-5 g/l



Release of dissolved material to the biosphere. U_o=0.03 l/m^2,year $C_{solub,U,ox}$=0.36 g/l migration distance = 5 m.

Figure 53	The fission products
Figure 54	The natural decay chains. Oxidizing migration conditions
Figure 55	The natural decay chains. Reducing migration conditions. $C_{solub,U,red}=1\cdot 10^{-5}$ g/l.



Release to the biosphere of dissolved material.
The fission products.
$U_0 = 0.03$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l. MIGRATION DISTANCE = 5 M.

Figure 53

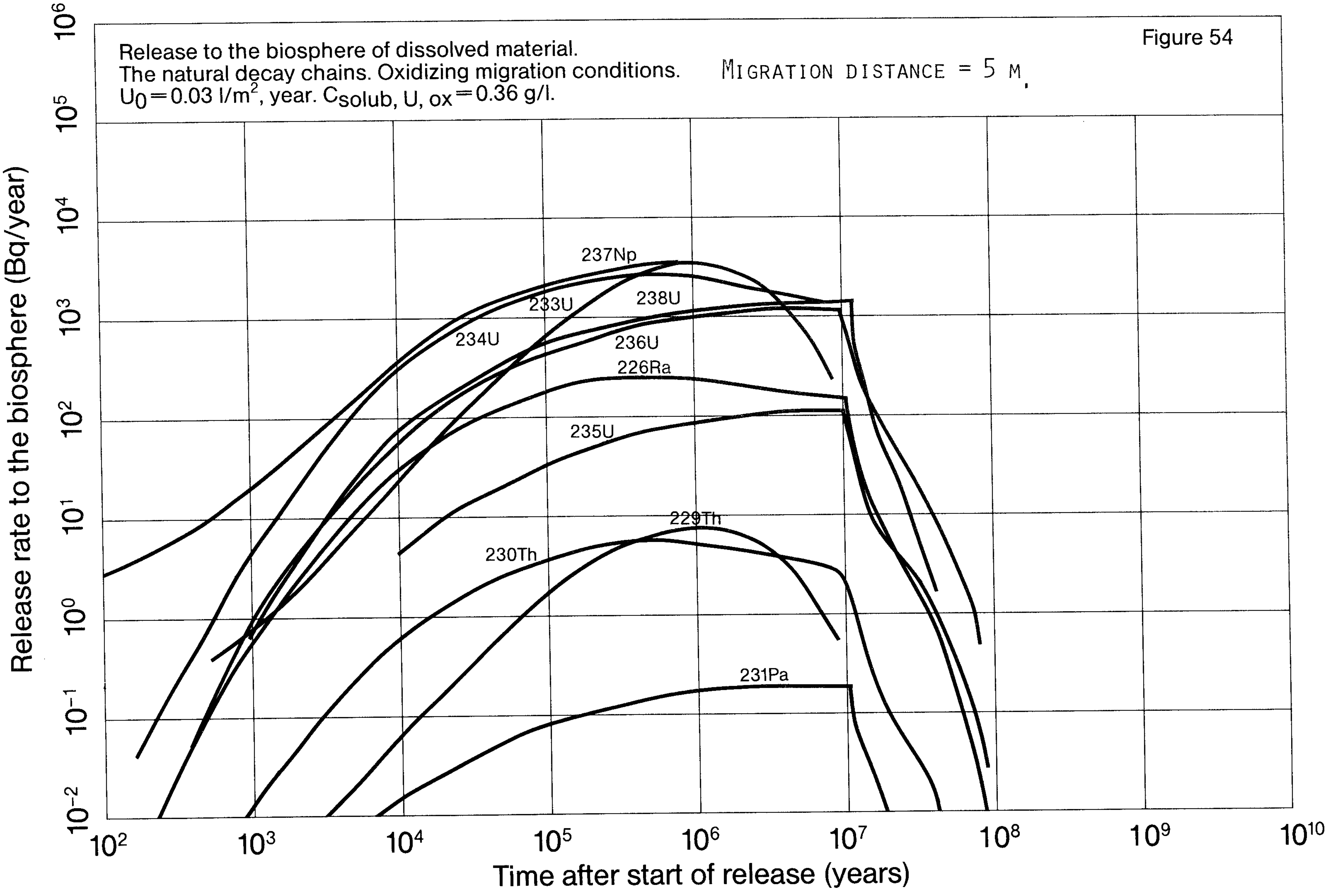
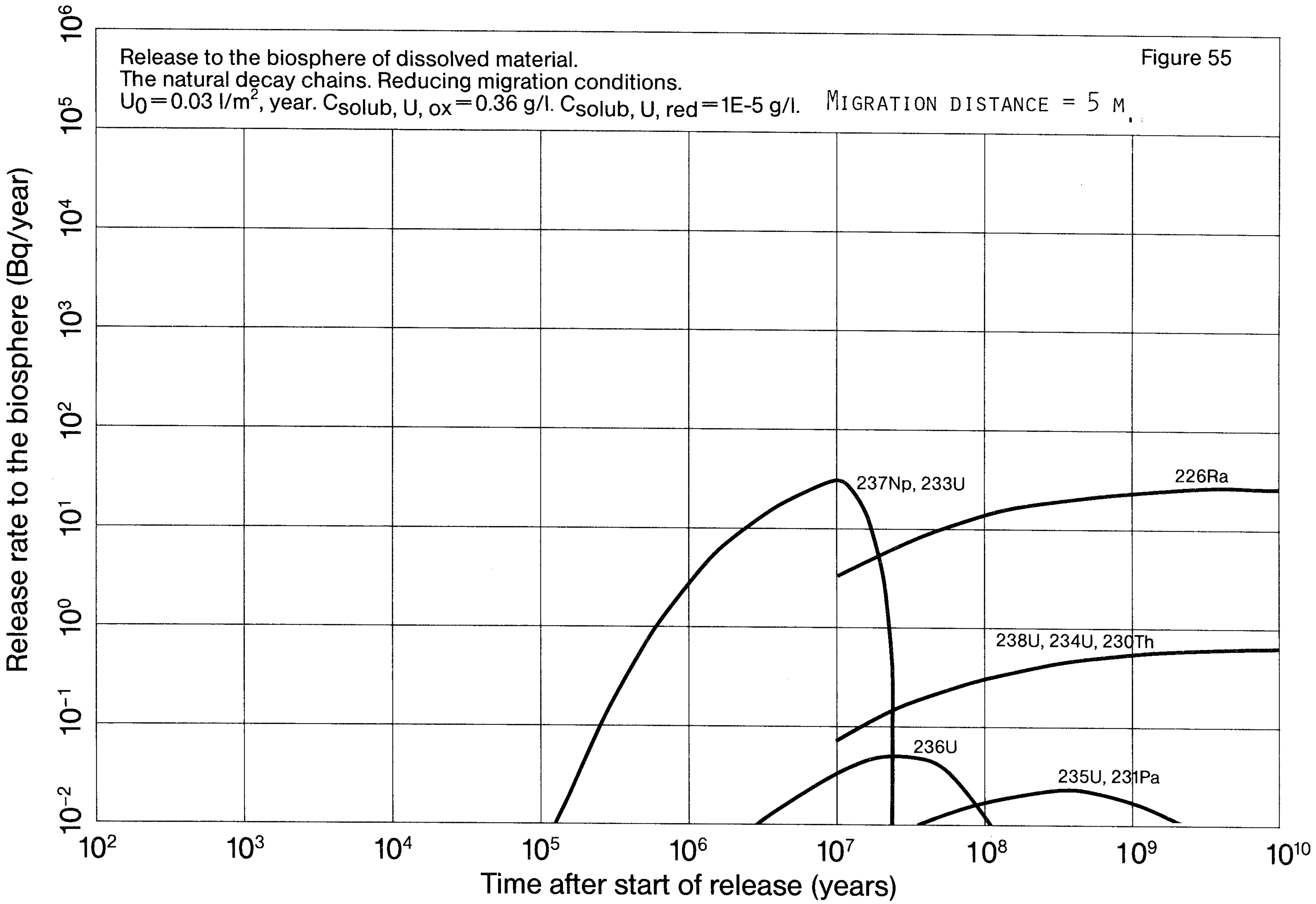


Figure 54

Release to the biosphere of dissolved material.
The natural decay chains. Oxidizing migration conditions. MIGRATION DISTANCE = 5 M.
$U_0 = 0.03$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l.


Release to the biosphere of dissolved material.
The natural decay chains. Reducing migration conditions.
$U_0 = 0.03$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l. $C_{solub, U, red} = 1E\text{-}5$ g/l. MIGRATION DISTANCE = 5 M.
Figure 55
Release rate to the biosphere (Bq/year)
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
Time after start of release (years)
10^2
10^3
10^4
10^5
10^6
10^7
10^8
10^9
10^{10}
237Np, 233U
226Ra
238U, 234U, 230Th
236U
235U, 231Pa

Release of colloidally bound material.
U_o=0.03 l/m^2,year $C_{solub,U,ox}$ =0.36 g/l.

Figure 56	The fission products except 99Tc
Figure 57	The natural decay chains and 99Tc from the oxidizing zone
Figure 58	The natural decay chains and 99Tc from the redox front. $C_{solub,U,red}=1 \cdot 10^{-5}$ g/l.

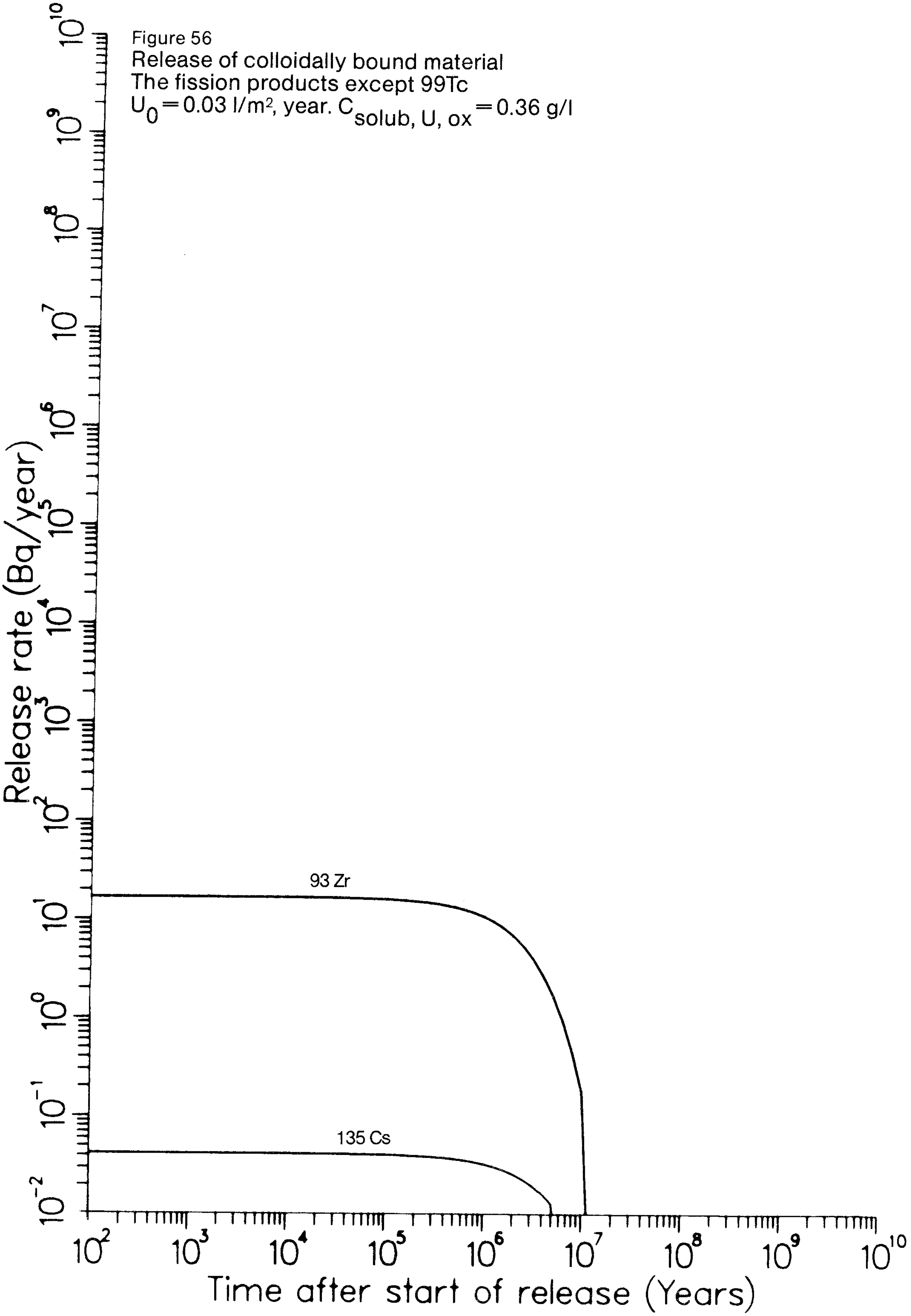


Figure 56
Release of colloidally bound material
The fission products except 99Tc
$U_0 = 0.03\ \mathrm{l/m^2}$, year. $C_{\mathrm{solub,\,U,\,ox}} = 0.36\ \mathrm{g/l}$

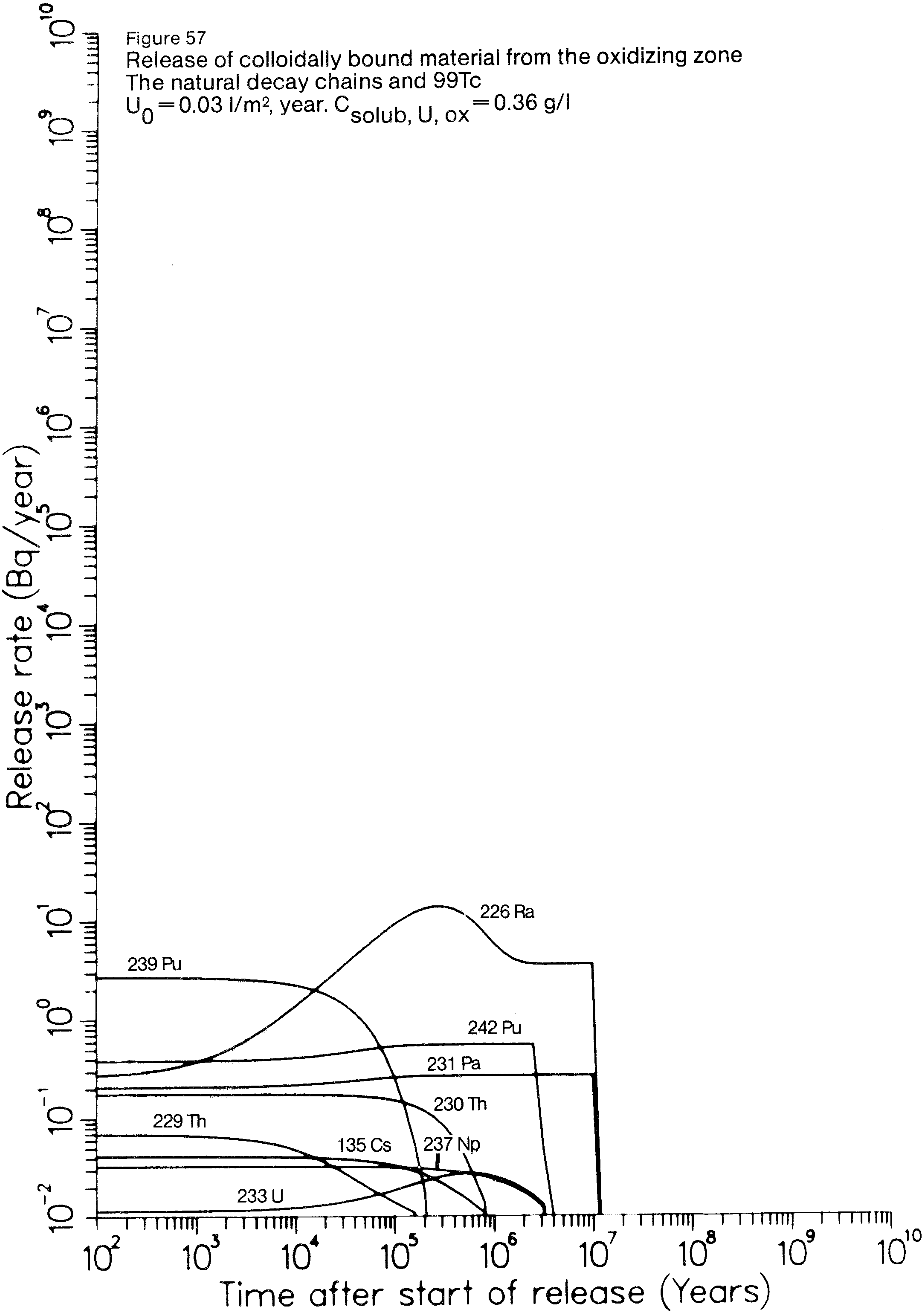


Figure 57
Release of colloidally bound material from the oxidizing zone
The natural decay chains and 99Tc
$U_0 = 0.03$ l/m², year. $C_{solub, U, ox} = 0.36$ g/l

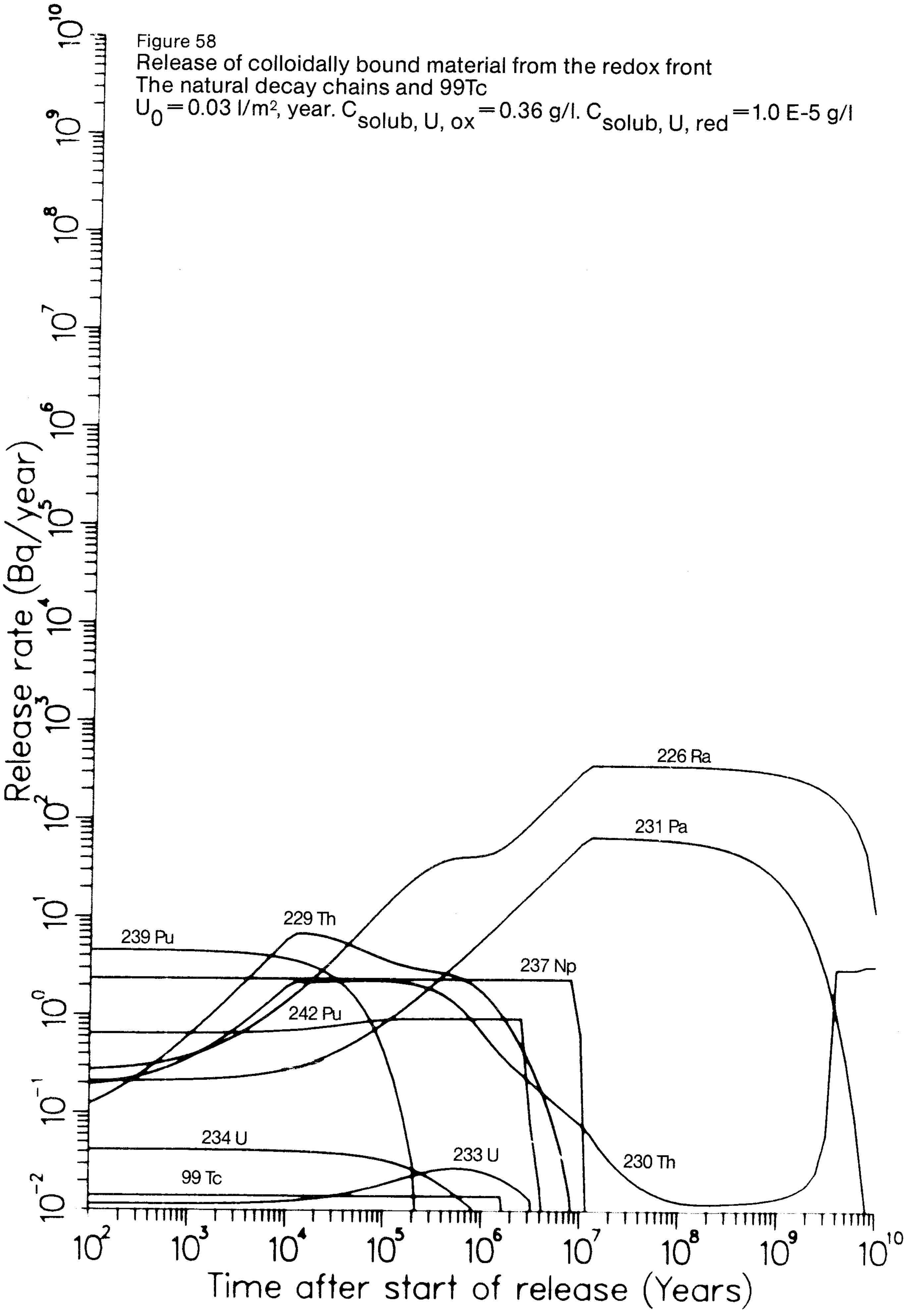


Figure 58
Release of colloidally bound material from the redox front
The natural decay chains and 99Tc
$U_0 = 0.03$ l/m^2, year. $C_{solub, U, ox} = 0.36$ g/l. $C_{solub, U, red} = 1.0$ E-5 g/l

5. THE EFFECT OF 10% OF THE IODINE AND CESIUM AVAILABLE IN PROMPTLY LEACHABLE FORM

5.1 Background

Iodine and cesium have been shown to migrate to the grain boundaries in the fuel during the burnup period.

This means that these substances to some extent are more readily leachable.

In the calculations below it is assumed that 10 % of the total content of iodine and cesium promptly dissolves the moment water reaches the fuel.
The mechanism thought to spread out the release of the dissolved material in time is a finite rate of displacement of the water volume which initially contains the dissolved material. This volume is taken to be the 1.7 m^3 of pore water residing in the clay buffer around a canister.

The primary release rate of promptly leachable material is shown in table 15.

TABLE 15 Primary Release rate of promptly leachable material (Bq/canister, year)
(In parenthesis the duration of the release in years)

Nuclide	U_0(l/m^2,year)	at t=10^2 years	at t=10^5 years
129I	0.03		3.5E4(5300)
	0.1	8.6E4(2100)	6.2E4(3000)
	1.0	3.0E5(630)	1.7E5(1100)
135Cs	0.03		3.7E5(5300)
	0.1	9.4E5(2100)	6.6E5(3000)
	1.0	3.2E6(630)	1.8E6(1100)
137Cs	0.03		
	0.1	3.0E10(2100)	
	1.0	9.9E10(630)	

5.2 Release from a whole repository

5.2.1 General

The collective effect of the release calculated above for a whole repository has been calculated on the following assumptions.

4400 canisters with one initially defective canister and an evenly distributed time for start of the leaching process for the rest of the canisters in the interval 10^5 to 10^6 years. This means that about 205 years will pass between each leach start.

For 0.03, 0.1 and 1.0 l/m^2, year groundwater flow rate this means that 26, 15 and 6 canisters will be leaching at the same time with the high rate calculated above.

5.2.2 Release to the geosphere of dissolved material

The release to the geosphere from the whole repository is shown in Table 16 below.

TABLE 16 Release to the geosphere of promptly leachable material (10%) from a repository consisting of 4400 canisters. (In parenthesis the maximum release from the other 90% residing in the fuel matrix calculated on the assumptions given elsewhere in this report)*.

Nuclide	U_0 (l/m^2,year)	Release The initially defect. canister (Bq/year,canister)	The other canisters (Bq/year,4400 can.)
129I	0.03		9.1E5(7.0E5)
	0.1	8.6E4(3.9E2)	9.3E5(1.2E6)
	1.0	3.0E5(3.5E3)	1.0E6(9.1E6)
135Cs	0.03		9.6E6(6.6E6)
	0.1	9.4E5(4.3E3)	9.9E6(1.2E7)
	1.0	3.2E6(3.9E4)	1.1E7(9.1E7)
137Cs	0.03		-
	0.1	3.0E10(1.3E8)	-
	1.0	9.9E10(1.2E9)	-

* Note: The different U_0 values also correspond to cases with different solubilities of uranium. The release rates shown above are not strictly additive. The effect on the maximum release rate to the biosphere will be very much dependent on the spread in time during the migration in the geosphere.

5.2.3 Release to the biosphere of dissolved material

For 129I the retention in the geological medium is very small and the figures given in table 16 are representative for the release to the biosphere.

For 135Cs the retention in the geological medium is sufficiently large to spread out the release from the promptly leachable material to such an extent as to make it indistinguishable from the release originating from the dissolution of the fuel matrix.

5.2.4 Release to the biosphere of colloidally bound material

The fraction of the release to the geosphere that will be transported to the biosphere by a colloidal mechanism will be:

$2.5 \cdot 10^{-5}$ for Cs and 0 for I.

5.3 Summary

The effect of more readily leachable material on the release rate to the biosphere will be none for Cs and a maximum twofold increase in the maximum release rate to the biosphere of I.

The increase in release rate for I can be expected to be further diminished by the more probable successive degradation of the copper canisters.

6. DISCUSSION

The leach rate of the spent fuel is determined by the equivalent flow rate of water dissolving and transporting the main matrix constituent UO_2. All other nuclides are released as the matrix dissolves away. The solubility of uranium in the water adjacent to the fuel is thus of prime importance. Due to the possible effects of alfa-radiolysis the water is assumed to be oxidizing and then uranium has a much higher solubility than in the naturally reducing waters in ferrous iron containing crystalline rock.

Some nuclides e.g. Pu and Th which have very low solubility will precipitate as the uranium dissolves away. Others e.g. Tc and Np which have very low solubility in reducing waters will precipitate at the redox front. For such nuclides the maximum concentration will be limited by their solubilities.

All dissolved nuclides will penetrate into the porous rock matrix by diffusion from the flowing water in the fissures. Most of the nuclides (except I and Sn) will sorb onto the inner surfaces of the pores or microfissures. This is by far the most important mechanism for retarding the nuclides in the present model. Compared to this sorption on the surfaces of the fissures with flowing water can be neglected.

Dispersion has been modelled as Fickian dispersion and high dispersivities have been used to ensure that early arrival of a portion of the nuclides is accounted for. (Sample calculations with small dispersion have shown to give smaller releases).

The treatment of colloidal transport by assuming that the nuclides are sorbed irreversibly on natural colloidal matter in the water is very conservative. If there were irreversible sorption mechanisms these would be active in the matrix also, effectively stopping a fraction of what now is modelled as reversibly sorbing nuclides.

In the present model calculations no account is taken of the retardation in the backfill. This is of little or no importance for the long lived canisters but would considerably retard some of the shorter lived nuclides released from a canister which fails initially.

Decrease of retardation by complex formation with fulvic and humic acids has been studied. 242 Pu was used as model nuclide. The release of this nuclide was increased but still did not become dominant. Other trivalent nuclides would be less important.

7. SYMBOLS USED

b	Half the fracture aperture	(m)
C	Concentration	(g/l)
D_p	Diffusivity in the pore water	$(m^2/year)$
D_L	Dispersion coefficient in the flowing water	$(m^2/year)$
K	Volume equilibrium constant $=K_d \rho_p$	(m^3/m^3)
K_d	Mass sorption coeffifient	(m^3/kg)
L	Migration length	(m)
M	Amount of a nuclide or substance	(kg)
N	Molar flux at the fracture/solid interface	$(mol/m^2,year)$
Pe	Peclet number $= u \cdot L/D_L$	
Q_{eq}	Equivalent water flow rate	(l/canister,year)
s	Spacing between fractures	(m)
t	Time	(years)
u	Linear velocity	(m/year)
U_0	Water flux in the undisturbed rock	$(l/m^2,year)$
z	Coordinate in the direction of the water flow	
ε_p	Porosity of the rock matrix	
ε_f	Flow porosity	
ρ	Density	(kg/m^3)
Φ	Mass flux	(g/canister,year)
λ	Decay constant	$(year^{-1})$

Subscripts:

coll	Colloidal particles
f	The fissure, the flowing water
i	Nuclide, isotope (Even used as a superscript)
j	Substance
ox	The oxidizing zone
p	The rock matrix
red	The reducing zone
solub	Solubility

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Royal Institute of Technology
KBS TR 83-71, may 1983

The Bateman equations

The amounts Ni of the respective nuclides in an isolated decay chain 1 2 3 can be written as:

$$N1= N01*exp(-L1*T) \qquad (1)$$

$$N2= N01*L1/(L2-L1)*(exp(-L1*T)-exp(-L2*T))+N02*exp(-L2*T) \qquad (2)$$

$$\begin{aligned} N3= {} & N01*L1*L2*(exp(-L1*T)/(L2-L1)/(L3-L1)+ \qquad (3) \\ & +exp(-L2*T)/(L_1-L_2)/(L3-L2)+ \\ & +exp(-L3*T)/(L_1-L_3)/(L2-L3)+ \\ & +N02*L2/(L3-L2)*(expL-L2*T)-exp(-L3*T))+ \\ & +N03*exp(-L3*T) \end{aligned}$$

Ni= amount of nuclide i (grams)
N0i= initial amount of nuclide i (grams)
T = time (years)
Li = decay factor for nuclide i $(years^{-1})$

APPENDIX B

Conversion of computer program TRUCHN from concentration to activity units

As TRUCHN works with absolute accuracy, one should strive to get the different peaks in a radionuclide chain at about the same heights. For certain chains with long-lived parent nuclides, like U-238 (U-234) Th-230 Ra-226, it is then advantageous to work in activity units instead of the original formulation in concentration units in TRUCHN. The conversion is done in the following way.

The migration of the ith member of a radionuclide chain is, in concentration units, given by (Rasmuson and Neretnieks, 1982).

$$\frac{\partial c_f^i}{\partial t} + u_f \frac{\partial c_f^i}{\partial z} - D_L \frac{\partial^2 c_f^i}{\partial z^2} = \alpha N^i - \lambda^i c_f^i + \lambda^{i-1} c_f^{i-1} \qquad (1)$$

$$K^i \frac{\partial c_p^i}{\partial t} = D_p \varepsilon_p \frac{\partial^2 c_p^i}{\partial r^2} + \frac{\beta}{r} \frac{\partial c_p^i}{\partial r} - K^i \lambda^i c_p^i + K^{i-1} \lambda^{i-1} c_p^{i-1} \qquad (2)$$

$$N^i = D_p \varepsilon_p \frac{\partial c_p^i}{\partial r} \quad r=\text{interface} \qquad (3)$$

Spherical blocks of rock matrix with a radius = r_0 means:

$$\alpha = -3(1-\varepsilon_f)/\varepsilon_f r_0 \qquad (4)$$

$$\beta = 2 \qquad (5)$$

To obtain the analogue formulation in activity units we use:

$$a^i = \lambda^i c^i \tag{6}$$

Substituting into equations (1)-(3) we obtain:

$$K^i \frac{\partial a_p^i}{\partial t} = D_p \varepsilon_p \left(\frac{\partial^2 a_p^i}{\partial r^2} + \frac{\beta}{r} \frac{\partial a_p^i}{\partial r} \right) - K^i \lambda^i a_p^i + K^{i-1} \lambda^i a_p^{i-1} \tag{7}$$

$$N^i = - D_p \varepsilon_p \left(\frac{\partial a_p^i}{\partial r} \right)_{r=\text{interface}} \tag{8}$$

$$\frac{\partial a_f^i}{\partial t} + u_f \frac{\partial a_f^i}{\partial z} - D_L \frac{\partial^2 a_f^i}{\partial z^2} = -\alpha N^i - \lambda^i a_f^i + \lambda^i a_f^{i-1} \tag{9}$$

Equations (7)-(9) are the equivalents of (1)-(3) in activity units. The only difference is in the last terms of equations (7) and (8) (build-up due to the decay of a parent nuclide). They include the factor ^{i} instead of $^{i-1}$.

The boundary conditions also have to be modified using equation (4). This is straight forward.

In the numerical code TRUCHN the modification (equations (7)-(9)) is done at the end of the main program HEART. The boundary conditions are appropriately modified in SUBROUTINE SURE.

Maximum release for an infinite source

Introduction:

To assess the importance of the primary source term to the geosphere migration calculation for a nuclide calculations have been performed on the stationary case with an infinite source i.e. a constant input concentration. The radioactive decay will establish a stationary concentration profile along the migration distance after a number of halflives for the nuclide studied.

An analytical expression has been derived for C/C_o as a function of the migration parameters and the halflife of the nuclide (Neretnieks et.al. 1983). The mathematical expression can be written in the following way:

$$\frac{C}{C_0} = \exp\{L\cdot(\frac{Pe}{2L} - ((\frac{Pe}{2L})^2 + \frac{\Psi}{D_L})^{\frac{1}{2}})\} \qquad (1)$$

$$\Psi = \lambda + \frac{\sqrt{\lambda\cdot D_e\cdot r_D}}{b} \qquad (2)$$

Where:

λ = the decay constant (years^{-1})

D_e = the effective diffusivity in the rock matrix = $D_p \varepsilon_p$ (m^2/year)

r_d = the retention factor = K_d rock

2b = fracture aperture (m)

Pe = the Peclet number = $u_f\ L/D_L$

D_L = the dispersion coefficient in the flow direction (m^2/year)

L = The migration length (m)

s = fracture spacing (m)

ε_f = flow porosity (m^2/m^2)

The case is derived for an infinite distance between fractures or in practical terms the fractures are so widely spaced that the concentration profiles from two fractures do not affect one another.

The penetration depths in the cases calculated for the KBS3 study ranges from millimeters to some centimeters for the nuclides notably affected by decay during the passage through the geologic medium. As this is a mere fraction of the 2.5 meters from the midline out to a fracture the diffusion process for the case with cubical blocks of rock matrix can effectively be modeled as diffusion into plane-parallell slabs.

To be able to compare with the solution of the numerical calculations, performed on qubical blocks of rock matrix with an edge length=S_{cube} and a fracture aperture=2b, the parameters b and D_L in (1) and(2) have to be modified.

For a medium with parallell fractures with a spacing=S and an aperture=2b, Snow (1968) has derived the expressions:

$$b \propto \sqrt[3]{S} \tag{3}$$

and

$$\varepsilon_f \propto \frac{2b}{S} \tag{4}$$

First the parameter ψ which is common to all the cases studied, is listed in table 1.

TABLE 1. Case independent characteristics

Nuclide	t(1/2)(years)	ψ (m^{-1})
H3	1.23+1	5.64-2
C14	5.73+3	1.21-4
Co60	5.27	6.15+1
Ni59	7.50+4	5.14-1
Ni63	1.00+2	1.41+1
Sr90	2.88+1	1.18+1
Zr93	1.53+6	5.09-1
Nh94	2.03+4	4.42
Tc99	2.14+5	9.63-3/1.52-1
I129	1.57+7	4.41-8
Cs135	2.95+6	4.10-2
Cs137	3.02+1	1.28+1
Sn126	1.00+5	5.93-6
Sm151	8.70+1	7.55+1
Eu154	8.5	2.42+2
Ho166m	1.2+3	6.65
Ra226	1.6+3	2.49
Th229	7.34+3	8.22
Th230	8.00+4	2.49
Th232	1.41+10	5.93-3
Pa231	3.28+4	3.89
U233	1.59+5	7.90-2/1.77
U234	2.45+5	6.36-2/1.42
U235	7.04+8	1.19-3/2.65-2
U236	2.34+7	6.51-3/1.46-1
U238	4.47+9	4.71-4/1.05-2
Np237	2.14+6	4.81-1
Pu238 red/ox(complex)	8.77+1	5.83+1/7.52+1(2.38+1)
Pu239 "	2.41+4	3.51/4.54(1.43)
Pu240 "	6.57+3	5.73/8.69(2.75)
Pu242 "	3.76+5	8.85-1/1.14(3.61-1)
Am241 (complex)	4.32+2	3.39+1(1.07+1)
Am242m "	1.52+2	5.71+1(1.81+1)
Am243 "	7.37+3	8.20(2.59)
Cm245	8.5+3	7.64
Cm246	4.7+3	1.03+1

Common characteristics for the geologic medium:

$D_e = 5 \cdot 10^{-14}\ m^2/s$
$b_{cube}\ /2 = 2.5 \cdot 10^{-4}\ m$
$s_{cube} = 5\ m$

The parameters special to each of the cases are shown in table 2. The C/C_0-values are shown in tables 3, 4 and 5.

To get the same volume to surface ratio as for the case with cubical blocks we will have to use

$$S = S_{cube}/3 \tag{5}$$

With a fixed Peclet number this means that

$$b=0.69 \cdot b_{cube} \tag{6}$$

$$D_z=D_{zcube}/2.08 \tag{7}$$

TABLE 2. Parameters characteristic to each of the cases

Case:	U_0=1 1/m, year Z=50m		U_0=0.3 1/m^2, year Z=100 m	U_0=0.1 1/m^2, year Z=100 m	U_0=0.03 1/m^2, year Z=5 m
Pe	2	50	2	2	2
$Pe/2_L (m^{-1})$	$2 \cdot 10^{-2}$	$5 \cdot 10^{-1}$	$1 \cdot 10^{-2}$	$1 \cdot 10^{-2}$	$2 \cdot 10^{-1}$
$D_L (m^2/year)$	1.2E2	4.81	7.26E1	2.40E1	3.61E-1
L(m)	50	50	100	100	5

TABLE 3. C/C_o for an infinite source. Values given for Pu and Am in parenthesis for K_d=0.5 m^3/kg

Nuclide	U_o=1 l/m^2,year	
	Z=50 m	
	Pe=2	Pe=50
3H	6.2E-1	5.6E-1
14C	1.0	1.0
60Co	7.6E-16	2.9E-68
59Ni	8.9E-2	7.7E-3
63Ni	9.5E-8	3.5E-96
90Sr	4.1E-7	1.4E-25
93Zr	9.0E-2	8.0E-3
94Nb	1.8E-4	2.4E-13
99Tc red/ox	9.1E-1/3.5E-1	9.0E-1/2.2E-1
129I	1.0	1.0
135Cs	7.0E-1	6.6E-1
137Cs	2.1E-7	6.4E-27
126Sn	1.0	1.0
151Sm	1.6E-17	1.4E-76
154Eu	3.9E-31	0
166mHo	2.0E-5	1.3E-17
226Ra	1.9E-3	6.8E-9
229Th	5.4E-6	2.9E-20
230Th	1.9E-3	6.8E-9
232Th	9.4E-1	9.4E-1
231Pa	3.2E-4	3.3E-12
233U red/ox	5.3E-1/5.8E-3	4.5E-1/6.1E-7
234U "	5.9E-1/1.1E-2	5.2E-1/6.7E-6
235U "	9.9E-1/7.8E-1	9.9E-1/7.6E-1
236U "	9.4E-1/3.6E-1	9.3E-1/2.3E-1
238U "	1.0 /9.7E-1	1.0 /9.6E-1
237Np "	9.8E-2	1.0E-2
238Pu "	2.0E-15/1.7E-17(5.7E-10)	3.0E-66/2.1E-76(2.2E-39)
239Pu "	5.0E-4/1.5E-4(1.1E-2)	2.3E-11/1.4E-13(6.2E-6)
240Pu "	4.7E-5/3.8E-6(1.3E-3)	6.1E-16/5.2E-21(1.5E-9)
242Pu "	3.3E-2/1.9E-2(1.5E-1)	3.6E-4/5.1E-5(3.0E-2)
241Am	7.7E-12(8.6E-7)	1.6E-48(5.0E-24)
242mAm	2.8E-15(9.8E-9)	1.8E-65(2.3E-33)
243Am	5.5E-6(1.6E-3)	3.1E-20(3.8E-9)
245Cm	8.7E-6	2.6E-19
246Cm	1.1E-6	1.9E-23

TABLE 4. C/C_o for an infinite source. Values given for Pu and Am in parenthesis for K_d=0.5 m^3/kg

Nuclide	U_o=0.3 l/m^2, year Z=100 m Pe=2
3H	1.4E-1
14C	9.9E-1
60Co	2.9E-40
59Ni	5.7E-4
63Ni	2.0E-19
90Sr	8.3E-18
93Zr	5.9E-4
94Nb	5.1E-11
99Tc red/ox	5.9E-1/2.5E-2
129I	1.0
135Cs	2.1E-1
137Cs	1.6E-18
126Sn	1.0
151Sm	1.4E-44
154Eu	1.4E-79
166mHo	1.9E-13
226Ra	2.4E-8
229Th	6.5E-15
230Th	2.4E-8
232Th	7.1E-1
231Pa	2.4E-10
233U red/ox	8.7E-2/4.4E-7
234U "	1.2E-1/2.2E-6
235U "	9.2E-1/3.1E-1
236U "	6.9E-1/2.7E-2
238U "	9.7E-1/2.7E-1
237Np "	7.5E-4
238Pu "	3.3E-39/1.7E-44(3.7E-25)
239Pu "	7.5E-10/3.7E-11(2.1E-6)
240Pu "	1.7E-12/2.5-15(9.3E-9)
242Pu "	4.2E-5/9.4E-6(2.2E-3)
241Am	5.7E-30(5.7E-17)
242mAm	8.3E-39(5.6E-22)
243Am	6.8E-15(1.7E-8)
245Cm	2.2E-14
246Cm	1.2E-16

TABLE 5. C/C_o for an infinite source. Values given in parenthesis for Pu and Am are for K_d=0.5 m^3/kg

Nuclide	U_o=0.1 $1/m^2$,year Z=100 m Pe = 2	U_o=0.03 $1/m^2$,year Z=5 m Pe=2
3H	1.9E-2	3.0E-1
14C	9.8E-1	1.0
60Co	8.2E-70	1.2E-28
59Ni	1.2E-6	6.4E-3
63Ni	1.4E-33	7.2E-14
90Sr	9.5E-31	1.0E-12
93Zr	1.2E-6	6.6E-3
94Nb	6.2E-19	6.7E-8
99Tc	2.9E-1/8.9E-4	7.5E-1/9.1E-2
129I	1.0	1.0
135Cs	3.9E-2	3.8E-1
137Cs	5.2E-32	3.1E-13
126Sn	1.0	1.0
151Sm	2.5E-77	1.1E-31
154Eu	0	0
166mHo	3.7E-23	1.3E-9
226Ra	2.7E-14	5.2E-6
229Th	1.0E-25	1.2E-10
230Th	2.7E-14	5.2E-6
232Th	4.2E-1	8.3E-1
231Pa	8.8E-18	2.0E-7
233U	8.0E-3/4.3E-12	2.1E-1/4.0E-5
234U	1.4E-2/7.3E-11	2.7E-1/1.3E-4
235U	8.0E-1/8.5E-2	9.6E-1/5.0E-1
236U	4.0E-1/1.0E-3	8.2E-1/9.7E-2
238U	9.1E-1/2.7E-1	9.8E-1/7.3E-1
237Np	4.9E-6	7.8E-3
238Pu	5.6E-68/3.6E-77(1.5E-43)	6.8E-28/1.2E-31(6.3E-18)
239Pu	6.6E-17/3.5E-19(6.7E-11)	4.5E-7/5.3E-8(1.2E-4)
240Pu	1.6E-2/2.0E-26(5.3E-15)	5.9E-9/5.9E-11(2.7E-6)
242Pu	1.2E-8/9.1E-10(1.2E-5)	1.0E-3/3.6E-4(1.7E-2)
241Am	6.6E-52(2.7E-29)	2.4E-21(4.0E-12)
242mAm	2.8E-67(5.2E-38)	1.3E-27(1.1E-15)
243Am	1.1E-25(1.4E-14)	1.2E-10(4.0E-6)
245Cm	8.5E-25	2.7E-10
246Cm	9.5E-29	6.7E-12

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